



THE THERMOPHYSICAL PROPERTIES OF  
**METALLIC LIQUIDS**

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Volume 1: **Fundamentals**

**TAKAMICHI IIDA & RODERICK I.L. GUTHRIE**

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# The Thermophysical Properties of Metallic Liquids

## Volume 1: Fundamentals

Takamichi Iida

*Professor Emeritus of Materials Science and Processing  
Osaka University, Japan*

Roderick I.L. Guthrie

*Macdonald Professor of Metallurgy at McGill University, and  
Director and co-founder of the McGill Metals Processing Centre  
McGill University, Montreal, Canada*

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# Preface

High temperature materials processing, such as smelting, refining, casting, welding, crystal growth, melt spinning, zone melting, and spraying, is an extremely complex area in which many thermophysical properties of metallic liquids are intimately involved. Numerous studies have been made to unravel this complexity on the basis of thermodynamics, reaction kinetics, and hydrodynamic analyses.

Between the 1950s and 1980s, great progress has been made in the industrial technology and science of process metallurgy, particularly for iron- and steelmaking processes. The advances in the science of extractive metallurgy have been described in numerous books, such as *Physical Chemistry of Metals* by Darken and Gurry (1953) and *Physical Chemistry of Melts in Metallurgy* by Richardson (1974). Advances in science and technology relating to liquid metallic processing operations are covered by books such as *Physical Chemistry of High Temperature Technology* by Turkdogan (1980) and *Engineering in Process Metallurgy* by Guthrie (1989). These books cover a wide range of topics relating to the physical chemistry of melts (i.e. liquid metals, salts, and slags): structure, physical properties, thermodynamic properties, reaction kinetics, interfacial phenomena, related mass/heat transport phenomena, and attendant fluid flows. As such, they are of great significance for process metallurgists. However, the former books place an emphasis on chemical, and reaction kinetics, while the latter emphasize process kinetics in metallurgical systems, related mainly to iron- and steelmaking. For such macroscopic or continuum treatments, the values of thermophysical properties are generally taken as empirical constants.

The advent of semiconductors, metallic glasses, and other functional materials since the late 1950s, and ever-increasing demands for the manufacture of high quality metallic materials have broadened the scope of high temperature science and technology of metallic liquids. At the same time, metallurgy has expanded into materials science and materials engineering. As such, materials process science has come to be based not only on chemistry but also on physics. This is especially true for the efficient manufacture of high quality metallic materials, where a detailed knowledge of the thermophysical properties of metallic liquids in question, based on the theory of liquids, is required. This atomistic, or microscopic, approach to materials processing, in contrast to equilibrium chemical thermodynamics, and process kinetics, provides an essential underpinning to understanding and interpretation of various phenomena in liquid metallic processing operations. From this viewpoint, the present authors published a technical book *The Physical Properties of Liquid Metals* (1988). The purpose of that book was to introduce theoretical equations based on structure, semi-empirical (semi-theoretical) equations, empirical equations, together with methods of experimental measurement and

experimental data, for the physical properties of liquid metals. This book was written for metallurgical and materials research workers.

Since the 1970s, there has been renewed interest in the thermophysical properties of almost all liquid metallic elements or simple metallic substances (i.e. liquid metals, semimetals, and semiconductors), following the advent of mathematical modelling techniques supported by powerful computers. Nowadays, computer simulation studies of materials processing operations, based on mathematical models, are widely used as a very useful tool for improving liquid metal and liquid metallic processing operations and product quality. Naturally, accurate and reliable data for the thermophysical properties of metallic liquids are indispensable, not only for the execution of computer simulations and for the development of mathematical models, but also for the direct solution of industrial high temperature processing operations. In particular, accurate data for almost all liquid metallic elements are first needed. To elaborate further on this matter, while multicomponent alloys are typically treated in liquid metallic processing operations, accurate and reliable data for the respective pure components of an alloy system are first needed as a starting point. In addition, we must go deeply into a study of an element's (i.e. simple substance) properties, in order to clearly understand the essence of a metallic liquid's thermophysical properties. For example, in the liquid state, the motions of atoms through the liquid are impeded by frictional forces set up by their nearest neighbours. Thus, the atomic transport coefficients, i.e. molecular viscosity and diffusivity, of metallic liquids are dominated by the frictional forces among atoms. Furthermore, the atomic transport coefficients cannot be entirely formulated in terms of thermodynamic properties alone, because the coefficients involve the movement of atoms. Owing to this, model theories, such as the hole theory or the rate process (activated state) theory, are unlikely to provide fruitful predictions of transport coefficients of metallic liquids.

A large number of research articles on the thermophysical properties of metallic liquids have been published in the last quarter century or so. Their results or main points have been organized in numerous review articles and books such as *Measurement and Estimation of Physical Properties of Metals at High Temperatures* by Mills in *Fundamentals of Metallurgy* edited by Seetharaman (2005). This book emphasizes the need for reliable data for the thermophysical properties of liquid materials (e.g. liquid metals) involved in high temperature processes. Mills states that thermophysical property data are beneficial in two ways: (1) in the direct solution of industrial problems, and (2) as input data for the mathematical modelling of processes. Unfortunately, the explanations for the various thermophysical properties of melts are condensed into 70 pages in that book, so details for each property are not described. (However, over 170 important articles are referred.)

As mentioned previously, there is an urgent need for reliable data for the thermophysical properties of metallic liquids. Indeed, accurate and reliable data for the thermophysical properties, even of liquid metallic elements, are still not necessarily plentiful. The absence of accurate data for metallic liquids is mainly due to the experimental difficulties in obtaining accurate values for these thermophysical properties at high temperatures. The measurements of the thermophysical properties of metallic liquids at high temperatures are time-consuming, expensive, and in some cases, impossible. Further, they require considerable experimental and metrological expertise. Thus, much effort

has been directed towards the development of reliable models for accurate predictions of the thermophysical properties of metallic liquids. Even so, for many years, little progress has been made on reliable models for accurate predictions. Early in this century, a new approach to making such predictions was presented for several thermophysical properties of all liquid metallic elements.

As seen from this brief historical outline (presented above), describing scientific approaches to the manufacture of metallic materials, the purposes of materials process science in this area are (1) to provide a clear understanding of the structure and thermophysical properties of metallic liquids based on the liquid state physics, and (2) to develop models that can be used for accurate predictions of the thermophysical properties of metallic liquids, particularly for all, or almost all, liquid metallic elements. In the area of materials process science, both accuracy and universality are required of any model for predicting the thermophysical properties of metallic liquids. Other purposes of materials process science are (3) to provide appropriate evaluation of the thermophysical property data for metallic liquids, and (4) to act as a guide to creation of new types of materials. In order to achieve these four main purposes, materials process science must incorporate a blending in of knowledge in many subject area, including process metallurgy, materials science, condensed matter physics (particularly, liquid state physics), molecular physics, chemistry (particularly, theoretical chemistry, and inorganic chemistry), and finally metrology (particularly for high temperature experiments on the thermophysical properties of metallic liquids).

The present book is divided into two volumes. This book of two volumes is a completely revised version of the authors' previous book *The Physical Properties of Liquid Metals* (1988): the present book lays emphasis on both the *Fundamentals (Volume 1)* and on *Predictive Models (Volume 2)* for accurate predictions of the thermophysical properties of metallic liquids. The performances of models/equations for several thermophysical properties (e.g. sound velocity, surface tension, viscosity) of liquid metallic elements are quantitatively assessed by determining relative differences between the calculated and experimental property values.

Volume 1 is intended as an introductory text explaining the structure and thermophysical properties of metallic liquids for students of materials science and engineering, and also for research scientists and engineers who have an interest in liquid metal and liquid metallic processing. The authors have tried to give simple explanations. However, the level of its contents is necessarily relatively advanced, since knowledge of the various scientific fields, mentioned above, is blended into one.

It comprises nine chapters, i.e. Chapters 1–9. Several basic matters for understanding the thermophysical properties of metallic liquids and for developing reliable models to accurately predict their thermophysical properties, together with methods for assessment of models/equations, are briefly described in Chapter 1. Chapter 2 is an introduction to the structure of metallic liquids. Two fundamental quantities in the theory of liquids, i.e. the pair distribution function and the pair potential are described. In particular, a reasonably detailed description of the distribution function is presented, because an understanding of the thermophysical properties of liquids must be based on a fundamental understanding of a liquid's atomic arrangement. Chapter 3 is concerned with the



density of metallic liquids. Although density, or number density (i.e. number of atoms per unit volume), is an indispensable and basic quantity, accurate data are not plentiful. A new model for the temperature dependence of liquid metallic element density (or volume expansivities) is introduced. Thermodynamic properties of a metallic liquid, i.e. evaporation enthalpy, vapour pressure, heat capacity, are outlined in Chapter 4. The velocity of sound in a liquid metallic element is described in Chapter 5. Dimensionless new common parameters for better predictions of several important thermophysical properties of metallic liquids can be extracted from their sound velocity data. The new common parameters give an indication of an atom's hardness or softness, as well as a piece of useful information about the structure of metallic liquids. The new parameters are also useful in discussions of anharmonic effects of atomic motions in metallic liquids. This theme is further developed in Chapter 6, dealing with surface tension, and Chapter 7, which covers the dynamic property, or transport property, viscosity, (also considered in volume 2). In Chapter 6, characteristic features of experimental data for metallic liquid surface tensions are identified. Reasons for large discrepancies among experimental data for metallic liquid viscosities are clarified in Chapter 7. In Chapters 6 and 7, the relationship between surface tension and viscosity for liquid metallic elements is discussed in some detail. Chapter 8 is concerned with diffusion in metallic liquids. Knowledge of diffusion is needed for many fields of engineering. However, even self-diffusivity data are extremely scarce; a predictive model for metallic liquid self-diffusivity, expressed in terms of well-known physical quantities, is presented. Metallic liquids, like solid metals, are characterized by high electrical and thermal conductivities. Fundamentals of electronic transport properties, i.e. electrical conductivity, or electrical resistivity, and thermal conductivity, of metallic liquids are discussed in Chapter 9, the final chapter. The essential points of methods for measuring density, surface tension, viscosity, diffusivity, electrical resistivity, and thermal conductivity are also described.

Volume 2 is designed for research scientists and engineers engaged in liquid metallic processing. In Volume 2, using as a basis the fundamental issues presented in Volume 1, we discuss models used for predicting accurate values of metallic liquid thermophysical properties.

It contains eight chapters, a glossary, and ten appendices. Essential points in building reliable models for accurate predictions of the thermophysical properties of liquid metallic elements are outlined in Chapter 10, from the standpoint of materials process science. Chapter 11 is devoted to the velocity of sound in liquid metallic elements. Useful dimensionless common parameters (or dimensionless numbers), which characterize the metallic liquid state, can be revealed through data for the velocity of sound. These common parameters allow for better predictions of several thermophysical properties of liquid metallic elements. Models, in terms of the common parameters, are discussed for the volume expansivity, evaporation enthalpy, surface tension, viscosity, and self-diffusivity of liquid metallic elements, in Chapters 12–16. The performances of the various models are evaluated by comparing them against experimental values (provided experimental data are available). Predicted, or calculated, data for sound velocity, volume expansivity, evaporation enthalpy, surface tension, viscosity, and self-diffusivity of liquid metallic elements are given in Chapters 11–16. In Chapters 10, 11, and 13–16,

atomic periodicity in values of each thermophysical property, discussed in this book, is illustrated for a large number of liquid metallic elements. In Chapter 17, the final chapter, a large number of experimental data for the physical quantities and the thermophysical properties of liquid metallic elements are compiled, although we emphasize that this book is not primarily a reference data book.

Prior to the Appendices, a glossary and/or supplementary explanations are provided. The periodic table contains many pieces of useful information about the thermophysical properties of elements; one form of periodic table is given in Appendix 1. Appendices 2 and 3 provide numerical expressions for determining the minimum values of relative standard deviation. SI units, unit conversions, fundamental physical constants in SI units, and the Greek alphabet, are given in Appendices 4–7. Occam’s razor is an essential guide to the development of any model for accurate predictions of the properties of materials; Appendix 8 cites Occam’s razor. In Appendices 9 and 10, calculated values of isothermal compressibility, structure factor, and the ratio of heat capacity of some liquid metallic elements at their melting point temperatures, are all listed.

Over the last half century, a huge number of research articles and review articles on the thermophysical properties of metallic liquids have been reported. Even so, our present knowledge of the thermophysical properties of metallic liquids is still lacking from the materials process science and engineering points of view. Accurate and reliable data for the thermophysical properties of metallic liquids are still not plentiful. Systematic investigations based on theory and experiment are greatly needed from the standpoint of materials process science and engineering. As such, we hope that this two volume book will not only be used for obtaining the relevant constants for the properties of specific liquid metallic elements, but will also help the user recognize the continuum between the microscopic and macroscopic approaches to liquid metallic processing operations. For the future, we hope this book will help in enabling great progress in materials process science.

Takamichi Iida  
Roderick I.L. Guthrie  
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# Principal Symbols

Numbers in parentheses refer to equations.

## Capital Italic

$A$	area (molar surface, oscillating plate, etc.)
$A$	parameter (2.11)
$A, B, C, D$	constants
$A_i$	area of component $i$
$A_r$	relative atomic mass
$A^*$	reduced area
$B_s$	isentropic, or adiabatic, bulk modulus
$C_A$	Andrade coefficient
$C_{Aw}(\eta)$	correction factor (8.13)
$C_{el}$	heat capacity of electron gas
$C_L$	constant (1.16)
$C_P, C_V$	heat capacity at constant pressure, at constant volume
$D$	self-diffusivity
$D_{HS}$	self-diffusivity in the hard-sphere fluid
$D_i, D_{S,M}$	solute, solvent (or base metal) diffusivity
$D_0$	frequency factor (8.21)
$D^*$	reduced rectilinear diameter (3.4), parameter (corresponding -state principle (8.17))
$E, E$	kinetic energy; electric field
$E, E_a$	resonant amplitude of plate in liquid, in air
$E_C^0$	molar cohesive energy (solid) at 0 K at 1 atm (101.325 kPa)
$E_F$	Fermi energy
$E_V^*$	height of potential barrier
$F$	Helmholtz free energy
$G$	Gibbs free energy
$G_s$	total molar surface energy
$\tilde{G}(Q)$	Fourier transform of $\{g(r) - 1\}$
$H$	enthalpy; height of liquid sample
$H^E$	enthalpy of mixing
$H_\mu, H_D$	constants or parameters (apparent activation energy for viscous flows or for diffusion)
$1/H$	shape factor of liquid drop

$\Delta_s^1 H_m$	enthalpy of melting
$\Delta_l^g H_b$	enthalpy of evaporation at the boiling point temperature
$\Delta_s^g H_0$	enthalpy of sublimation at 0 K
$I$	intensity (of X-ray beam); moment of inertia of suspended system
$\mathcal{J}$	flux of matter
$K$	coverage independent adsorption coefficient
$K, L, M, \dots$	shells (energy level)
$K, K_0$	apparatus constants
$K_f$	force constant
$M$	molar mass
$N$	number of atoms; number of samples
$N_A$	Avogadro constant
$P$	pressure
$\Delta P$	pressure difference
$P(T)$	probability function
$P(A), P(B)$	proportion of vibrator $A, B$ (7.38)
$P_1$	Legendre polynomial
$P_m$	maximum bubble (gas) pressure
$Q$	parameter (6.11)
$Q_E$	activation energy (8.21)
$R$	molar gas constant; radius
$S$	entropy
$S, S(N)$	relative standard deviation
$S_A$	surface area
$S(Q)$	structure factor, or interference function
$S_{\alpha\beta}(Q)$	partial structure factor
$S_S$	molar excess surface entropy
$\Delta_s^1 S_m$	entropy of melting
$\Delta_l^g S_b$	entropy of evaporation at the boiling point temperature
$T$	absolute temperature; period of oscillation
$T^*$	reduced temperature
$U$	sound velocity; total internal energy
$U_b$	mobility
$U_1$	total internal energy (liquid)
$U(Q)$	pseudopotential
$V$	volume; molar volume (e.g. (7.14))
$V_{\text{mol}}$	molar volume
$V_A$	molar volume for a binary system
$V^E$	excess volume (3.23)
$\Delta V_m$	volume change on melting
$W(\phi)$	probability of a thermal fluctuation
$X, X', Y, Z, Z'$	parameters (measurements of surface tension)
$Z$	first coordination number; partition function (2.15)
$Z_i, Z_s$	first coordination number within bulk, at surface

**Lower Case Italic**

$a$	average interatomic distance
$a, b, c, d, q$	constants
$a_i, a_s$	activity of component $i$ or $s$
$c$	concentration
$d$	diameter of an atom (or ion); distance between solute and solvent ions
$dv$	volume element
$e$	electron (or electronic) charge
$f$	atomic scattering factor
$f$	activity coefficient; surface-packing, (or configuration), factor
$f, f_a$	resonant frequency in liquid, in air
$f(s), f_z$	interatomic forces
$g$	acceleration of gravity
$g(r)$	pair distribution function
$g_{\alpha,\beta}(r)$	partial pair distribution function
$g_s$	surface energy per unit area
$h$	depth of immersion
$h, \hbar$	Planck, Dirac constants ( $\hbar = h/2\pi$ )
$\bar{h}$	difference in height
$i$	chemical constant
$j$	electric current density
$k$	Boltzmann constant
$k_F$	radius of the Fermi sphere, or wave (number) vector
$k_f$	force constant
$k_0$	dimensionless numerical factor (6.37)
$l$	orbital quantum number
$l$	effective thickness of interface; length; mean free path (electron)
$m$	atomic mass (or mass of an atom); mass of a liquid drop
$m$	magnetic quantum number
$m_e$	electron mass
$n$	principal quantum number; repulsive exponent
$n$	amount of substance (mole in SI units)
$n$	number density; electron number density (9.3)
$n_0$	average number density
$\Delta n$	fluctuation in number density
$\dot{q}$	power input (9.15)
$r$	radial distance; radius
$r_0$	value of $r$ at the left-hand edge of the first peak in $g(r)$ curve
$r_m$	value of $r$ at the first peak, or the main peak, in $g(r)$ curve
r.d.f.	radial distribution function
$s$	spin quantum number
$s$	surface tension correction (3.16); distance of displacement
$s, d, p, f$	block (cf. Table 1.7)

$s, p, d, f \dots$	subshells (energy level)
$t$	time
$u_v$	internal energy (during vibration) (1.14)
$u, v$	pairwise interaction energy among atoms within bulk, on surface
$v$	volume of immersed suspension wire (3.16)
$v$	average electron velocity (9.11)
$v_0$	close-packed molecular volume
$v_F$	Fermi velocity
$v_f$	average free volume per molecule
$w$	work, or energy, necessary to separate an atom
$\Delta w$	apparent loss of weight
$x$	molar, (or mole), fraction; length
$x, y, z$	coordinate axes
$z$	number of valence electrons
$z^E$	excess valence

## Greek

$\alpha$	volume expansivity
$\alpha_a$	absorption coefficient
$\underline{\alpha}$	parameter related to the distance over which the interatomic force extends
$\alpha, \beta$	phases
$\beta$	correction factor (for Lindemann's equation)
$\beta_S$	coefficient of sliding friction
$\Gamma$	damping constant
$\Gamma_s$	excess surface concentration of solute $s$
$\Gamma_s^0$	saturation coverage by solute $s$
$\gamma$	surface tension
$\gamma_G, \gamma_{G,E}, \gamma_{G,T}$	Grüneisen constants (cf. Section 5.5)
$\gamma_h$	$\equiv C_P / C_V$ (ratio of the isobaric and isochoric heat capacities)
$\gamma_M$	surface tension of mixtures
$\gamma_0$	surface tension of pure solvent (6.58)
$\gamma_0$	$\equiv k_0 C_A$
$\Delta$	dimensionless variable (3.6)
$\Delta, \Delta(N)$	global delta (1.21)
$\delta$	amplitude of vibration of each atom (1.12)
$\delta, \delta_0$	logarithmic decrements
$\delta_i$	relative difference between experimental and calculated values for $\chi_i$
$\zeta_A$	parameter (8.20)
$\zeta_f$	friction coefficient
$\zeta_H, \zeta_S, \zeta_{SH}$	friction coefficients due to hard-core collision, to soft interaction, to cross effect
$\eta$	packing fraction (or packing density)
$\eta_l$	phase shift

$\theta$	Einstein characteristic frequency
$\theta$	contact angle; angle
$2\theta$	scattering angle
$\theta_S$	fractional coverage
$\kappa$	transmission coefficient
$\kappa_S$	isentropic, or adiabatic, compressibility
$\kappa_T$	isothermal compressibility
$\Lambda$	temperature dependence of the density of liquid metallic elements
$\lambda$	thermal conductivity
$\lambda$	wavelength; screening radius (8.36)
$\mu$	viscosity; constant ( $\mu \equiv bT_c / \rho_c$ , (3.4))
$\mu_A$	viscosity of binary liquid mixtures (or alloys)
$\mu_\kappa, \mu_{\phi(\sigma)}$	viscosity due to kinetic contribution, to a hard-sphere collision
$\mu_\phi$	soft attractions
$\mu^E$	excess viscosity
$\nu$	kinematic viscosity ( $\nu \equiv \mu / \rho$ )
$\nu$	atomic frequency
$\xi_E, \xi_T, \xi$	dimensionless common parameters ( $\xi \equiv \xi_T / \xi_E$ )
$\pi$	pi (3.141592 . . .)
$\rho$	density
$\rho_A$	alloy density
$\rho_e$	electrical resistivity
$\sigma$	effective hard-sphere diameter; diameter of molecule (7.11)
$\sigma_e$	electrical conductivity
$\tau$	mean free time
$\Phi$	total potential energy
$\phi$	excess binding energy; fluctuation in kinetic energy (7.21)
$\phi_H, \phi_S$	hard-sphere potential, soft potential
$\phi(r)$	pair potential (energy)
$\tilde{\phi}_S(Q)$	Fourier transform of the long-range part of potential
$\chi_i$	thermophysical property (e.g. sound velocity, surface tension, viscosity, etc.)
$\omega_c$	characteristic oscillation frequency

## Subscripts

b	at boiling point
c	at critical point
m	at melting point; value of $r$ at the first peak, or the main peak, in $g(r)$
	curve
g	gas
l	liquid
s	solid



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