Strengths and Weaknesses of Predictive Methods for Estimating Thermophysical Properties

Thomas E. Daubert

Department of Chemical Engineering, The Pennsylvania State University, 165 Fenske Laboratory, University Park, Pennsylvania 16802

Twenty-five years of research on predicting properties are reviewed on the basis of projects resulting in the ongoing American Petroleum Institute's *Technical Data Book - Petroleum Refining* and the AIChE-DIPPR's *Data Prediction Manual* and *Data Compilation*. While methods at this time are accurate for some properties and certain classes of compounds and their mixtures, very little improvement is evident for other properties. While many weaknesses are directly attributable to a paucity of data available for deriving prediction methods, complexity of certain molecules and their interactions have yet defied development of useful prediction techniques. Properties to be discussed will be limited to liquids and vapors and will include basic thermophysical properties, derived thermal and equilibrium properties, and transport properties. Results from evaluation of predictive methods against experimental data will be used for assessment of the strengths and weaknesses of current methods. Suggestions for data necessary for development of improved predictors will be made. Properties to be discussed include density including *PVT* relations, vapor pressure from the triple to the critical point, critical properties, enthalpies and entropies of formation and phase transition, thermal properties, viscosity, thermal conductivity, diffusivity, and surface tension.

Introduction

Continuous work for the past 25 years in preparing the API's *Technical Data Book - Petroleum Refining* (extant 1994) and for the past 16 years in the AIChE-DIPPR projects leading to the *Manual for Predicting Chemical Process Design Data* (extant 1990) and the *Data Compilation* (extant 1995) have given much perspective on the gaps that occur in both experimental data availability and predictive method applicability, consistency, and accuracy.

In the 12 years that have passed since a previous paper on this subject (Daubert and Danner, 1983), it is disconcerting that only minimal progress has been made in resolving the data and prediction method deficiencies. The most recent review of the best methods for predicting thermophysical properties alluded to in this paper is the prediction and correlation of physical properties subsection of the forthcoming seventh edition of Perry's *Chemical Engineering Handbook* by Daubert and Buck (1996). Both limitations of the methods included as well as extensive references to previous work are given. A listing of the best methods of prediction of properties as determined for the AIChE-DIPPR Data Compilation Project are given by Daubert et al. (1990).

While experimental data are scarce for many properties, inhibiting derivation of prediction methods, the problem of deriving reasonable prediction methods is often compounded by the fact that molecules of interest today tend to be larger and more complex, making simple prediction techniques inaccurate. On the other hand amazingly simple techniques will often predict some important properties with only a minimum number of input parameters. However, very little data to test methods for the more complex molecules are being measured.

The advent of simulators, where almost anything can be estimated, makes it important that users understand the effects of poorly estimated thermophysical properties on their work. The idea that new data and models are no longer necessary is simply not true. Sandler (1994) speaks well to this issue and also points out that the largest problem in the use of simulators is with identifying the most accurate model to use for a specific situation.

This paper will attempt to point out both advances and roadblocks in our continuing efforts to develop methods to predict accurate thermophysical properties for industrial applications. No attempt will be made to discuss scientific deficiencies and needs. Treatment of this vast subject will necessarily be brief although appropriate references provide more detail. With overdesign rapidly becoming a thing of the past and with expensive experimental measurement of data limited to the most important properties, recommendations are made as to the most useful improvements in prediction methods needed at this time.

Prediction—Hierarchy of Properties

The status of any particular property in the hierarchy of properties is dependent on both the importance of (1) the use of the specific property by itself or as an input to calculation or prediction of another property and (2) the accuracy to which the property must be known for the ultimate purpose to which it is being put. An example of a property which in itself must be accurate is the vapor pressure of a fluid which is to be used in calculating the vapor−liquid equilibria between close boiling compounds for design of a distillation tower. In this case an error of only 1 or 2% may cause severe over- or underdesign, both unsatisfactory. Thus prediction methods for vapor pressure must be quite accurate. An example where the ultimate use of a property is important would be the liquid density as a function of temperature of a light hydrocarbon such as butane. If the density is being used to size a pump, an error of 1% in the liquid density will have a negligible effect. However, if the density is to be used for custody

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Table 1. Ranking of Properties

property	some primary uses
1. critical properties	phase behavior thermal properties vapor−liquid equilibria
2. vapor pressure	enthalpy of vaporization vapor−liquid equilibrium
3. vapor and liquid density	sizing and flow calculations
4. thermal properties	energy balances chemical reaction calculations
5. viscosity	flow of fluids and heat
6. thermal conductivity	flow of heat

transfer, an error of 1% could have major financial implications to the seller or the buyer. Thermal conductivity illustrates a property used for different purposes. As the liquid thermal conductivity of an organic fluid is primarily used to design heat exchangers and affects the calculation of heat transfer coefficients as one of several variables, an error of 10% is probably satisfactory. On the other hand the thermal conductivity of a liquid metal used as a conductor must be known to a much higher accuracy as the property is used directly in the conduction calculation.

Based on the above reasoning and limited primarily to organic (hydrocarbon and nonhydrocarbon) fluids, a ranking of basic properties of most importance is given in Table 1. No attempt is made to include every property of interest. Each of the pure component properties will be discussed in succeeding sections with respect to data availability and predictive method development. Reference will be made to past work as well as to more recent studies. Both defined mixtures and undefined mixtures such as petroleum fractions to which we have devoted much time and effort are generally not within the scope of this paper. Predictive techniques such as equations of state which utilize the basic properties as well as calculate derivative properties such as vapor−liquid equilibrium will only be mentioned briefly.

Analysis of Data and Predictive Technique Availability

Critical Properties. Experimental determination of critical properties by visual techniques has been limited to compounds which both do not decompose as the temperature approaches the critical point and do not have a critical temperature above a practical limit of about 350 °C—up to 10 carbon atom hydrocarbons and stable organics of similar molecular weight. Except for the work of Kay et al. for the American Petroleum Institute, few systematic studies were carried out except for the AIChE-DIPPR Project 851 measurements during the last 10 years. The sealed-tube method of Mogollon et al. (1982) has allowed determination of critical temperatures for unstable fluids such as in the work of Smith et al. (1987). A low residence time flow apparatus developed by Rosenthal and Teja (1989) has been useful for similar work. Ambrose and Young (1995) have reviewed the methods exhaustively, while Wilson et al. (1995) have reported their most recent results. While work in this area appears to be increasing, the total number of hydrocarbon and nonhydrocarbon organic compounds for which experimental data are available are only about 450 for critical temperature, 350 for critical pressure, and 250 for critical volume. (See Elliott and Daubert (Table 2) and Kallianpur et al. (Table 2)).

Prediction methods most utilized for critical properties of pure compounds have generally been of the first-order group contribution type of Lydersen (1955) and Ambrose (1978, 1979) and their modifications. Jalowka and Daubert (1986) and Daubert and Bartakovits (1989) published a second-order group contribution utilizing the group definitions of Benson (1976) which allows neighbor and next nearest neighbor groups to be taken into account, thus effectively differentiating among isomers. All of these methods only require molecular weight, molecular structure, and normal boiling point as input parameters. Continuing evaluation of these methods by O'Neil (1993) and Babcock (1995) indicate that while the second-order methods work somewhat better than the first-order methods for compounds with one functional organic group (e.g. ketones), these methods do not predict much more accurately than first-order methods for multifunctional compounds (e.g. keto alcohols). The amount of data for multifunctional compounds is insufficient to derive group contributions or corrections. Thus, continuing experimental work for such compounds is necessary if further progress is to be made.

For defined mixtures the critical temperature and pressure data base is quite extensive due to the long term efforts of Kay and his co-workers. The critical volume data available is quite small. Details of the data sets are given by Elliott and Daubert (1987) and Kallianpur et al. (Table 1). Accurate methods for prediction were limited to complex mixing rules such as those of Chueh and Prausnitz (1967) until use of equations of state to determine excess critical temperatures and pressures of hydrocarbon (including nonhydrocarbon gas) mixtures were found to be just as simple and accurate as the complex mixing rules. Elliott and Daubert (Table 2) discuss the use of the Soave equation in detail. An accurate equation of state method for prediction of critical properties of nonhydrocarbon mixtures has not yet been proposed.

The most significant gap in prediction methods for critical properties is for pure multifunctional compounds. As soon as sufficient experimental data become available, the second-order group contribution methods can be redeveloped. Such work is essential as we move toward calculations involving more and more complex chemicals. In this very important area, the structure for prediction exists for development as the experimental data base expands.

Vapor Pressure. Although the quantity of available vapor pressure data are quite large as reported by Daubert and Thorwart (Table 2) for hydrocarbons and Gupte et al. (Table 2) for nonhydrocarbon organics, certain families such as acetylenes, epoxides, and some sulfur compounds have little coverage. In addition a real dearth of data exists above a reduced temperature of 0.7 except for the common low molecular weight compounds. Experimental methods and possible compound decomposition, especially for multifunctional compounds at higher temperatures, has deterred such measurements.

For hydrocarbons, the Lee and Kesler (1975) method requiring critical temperature, critical pressure, and acentric factor as inputs is a very accurate predictor for pure compounds so long as at least the normal boiling point is known. For nonhydrocarbons, at least two and preferably three experimental vapor pressure points are required for the use of the accurate prediction method of Riedel (1954). The accuracy of the Riedel method is improved the wider the available experimental data range, although often the only available data are at pressures below atmospheric (such as 1, 10, and 20/100 mmHg), especially for higher boiling compounds.

Although many attempts have been made to predict vapor pressures from molecular parameters or from a single limiting experimental vapor pressure point, no method of sufficient accuracy has surfaced. Gupte and Daubert (1985b) advanced a generalized corresponding states model using the radius of gyration and the reduced dipole moment in addition to the critical point and normal boiling point as parameters. This method is superior to the Riedel method but requires more information. Generalized methods by family, dependent only on carbon number, were advanced by Ambrose and Sprake (1970) and refined by Dickson and Daubert (1988). Methods dependent only on molecular group contributions based on the reduced normal boiling point rather than the reduced critical temperature were attempted in this laboratory but exhibited errors higher than deemed appropriate for vapor pressure. A new group contribution method has been advanced by Tu (1994) for all organic compounds and shows some promise.

Liquid Density. The quality and quantity of saturated liquid density data for hydrocarbons is reasonable, though data tend to be for lower molecular weight compounds and do not normally extend above the normal boiling point. For nonhydrocarbon organics, the corresponding data base is limited to a few members of each homologous series. Compressed liquid densities are limited. Lobo et al. (Table 2) and Umesi et al. (Table 2) review the available data.

So long as at least one experimental density value is available for pure compounds, the modified Rackett method of Spencer and Danner (1972) predicts accurately for essentially all organics. No accurate method is available to predict the liquid density when no experimental value is available unless the compound is a member of a well-defined family. Efforts should be made to develop a second-order group contribution liquid density prediction method that will require no experimental liquid density data.

Vapor Density. While experimental values of vapor density are not readily available for most compounds, prediction by corresponding states methods are relatively accurate for both hydrocarbons and nonhydrocarbons. Selected analytical cubic equations of state are accurate for nonpolar compounds.

Thermal Properties. Thermal properties include (1) ideal gas heat capacity, entropy, and enthalpy, (2) enthalpy of vaporization, (3) liquid heat capacity, and (4) real gas and liquid enthalpy. Reports by Daubert et al. (Table 2) on hydrocarbons and Gupte et al. (Table 2) on nonhydrocarbons discuss this area in detail. Data for ideal gas properties are either calculated from spectroscopic data or predicted from structure. While nonhydrocarbon spectroscopic values are not plentiful, the prediction methods available such as those of Benson (1976) and Domalski and Hearing (1993) have been developed to accurately predict most compounds.

Experimental enthalpy of vaporization data are available for a large number of compounds but usually only are measured at a few temperatures, the normal boiling point being the most prevalent. For some families only one value exists. Several methods such as the method of Watson (1931) are available to expand the temperature range but a priori estimation is limited to compounds for which vapor pressure and vapor and liquid density data are available in the temperature range of interest so that the thermodynamically exact Clapeyron equation can be used. For nonhydrocarbon compounds a method using the Clapeyron equation with only the acentric factor, normal boiling point, the Rackett parameter, and the critical temperature and pressure developed by Gupte and Daubert (1985a) compares favorably to the best previous methods. Corresponding states methods are also reasonably accurate.

Liquid heat capacity data, except for common hydrocarbons, are often only available near room temperature. Prediction methods such as the methods of Ruzicka and Domalski (1993) and Chueh and Swanson (1973) and the Lee and Kesler (1975) Pitzer corresponding states method are convenient but often do not predict consistent values, especially above the normal boiling point. Thus, improvements in the methods are necessary.

Real gas and liquid enthalpy data are available for hydrocarbons and their mixtures but are almost nonexistent for nonhydrocarbons, other than gases. However, the Pitzer method is an excellent predictor for hydrocarbons and appears to be more accurate for nonhydrocarbons than other methods. However, without additional nonhydrocarbon experimental enthalpy or liquid heat capacity data, attempts to improve prediction methods would not be productive.

Viscosity. As discussed by Jain et al. (Table 2), Fitzgerald and Daubert (Table 2), and Bhethanabotla et al. (Table 2), low-pressure vapor and liquid viscosity data are reasonably plentiful for both hydrocarbons and nonhydrocarbons. Compressed pure component vapor data and liquid data above the normal boiling point are scarce. Vapor viscosities at low pressures are accurately predicted by several methods requiring primarily critical temperature, critical pressure, and molecular weight as inputs. Low-pressure liquid viscosity is most readily predicted from structure and first-order group contribution methods such as the method of van Velzen et al. (1972). The behavior of data for specific families of compounds as discussed by Ibrahim and Daubert (1989) can also be used for prediction purposes. For liquid viscosity at high pressures, corresponding states types methods have been used for hydrocarbons but no methods have been advanced for nonhydrocarbons.

Thermal Conductivity. As discussed by Crooks et al. (Table 2), Wu and Daubert (Table 2), and Nagvekar et al. (Table 2) the data base available for both pure vapor and liquid thermal conductivity is relatively small. Vapor phase pure component prediction methods are numerous and at least as accurate as the data available for hydrocarbons and some nonhydrocarbons. For pure liquid compounds the major methods of prediction require critical temperature, normal boiling point, and molecular structure. A second-order group contribution method, developed by Nagvekar and Daubert (1987), is completely general and only requires molecular structure and critical temperature as inputs.

Diffusivity. Both data and predictive methods were discussed by Umesi et al. (Table 2) and Northup et al. (Table 2). As data are scarce, prediction methods are not very accurate although usage does not require high accuracy.

Surface Tension. Engel et al. (Table 2) and High et al. (Table 2) reviewed both data and predictive methods. Available methods of prediction involving group contributions are sufficient.

Summary and Conclusions

The question of what is necessary for the development of improved prediction methods is the major subject of this paper. Data needs have been briefly noted throughout the paper and in the references. Earlier, the hierarchy of properties was given. Thus, this section will point out only the absolutely necessary data required after which a general framework of prediction is suggested.

Table 2. General References

title[a] (documentation of the basis for the selection of the contents of	co-authors	date
Chapter 4, Critical Properties	J. R. Elliott, T. E. Daubert	1984
Chapter 5, Vapor Pressure	T. E. Daubert, M. J. Thorwart	1993
Chapter 6, Density	J. P. Lobo, R. P. Danner, T. E. Daubert	1984
Chapter 7, Thermal Properties	T. E. Daubert, G. Singh, K. Hanawalt, J. Schnitzer	1991
Chapter 11, Viscosity	V. Jain, T. E. Daubert, R. P. Danner	1984
Chapter 11, Viscosity	D. J. Fitzgerald, T. E. Daubert	1996
Chapter 12, Thermal Conductivity	R. G. Crooks, T. E. Daubert, R. P. Danner	1980
Chapter 12, Thermal Conductivity	R. Wu, T. E. Daubert	1996
Chapter 13, Diffusivity	N. O. Umesi, R. P. Danner, T. E. Daubert	1980
Chapter 10, Surface and Interfacial Tension	M. J. Engel, T. E. Daubert, R. P. Danner	1982

title	co-authors	date
Chapter 2, Critical Properties	C. A. Kallianpur, T. E. Daubert, R. P. Danner	1982
Chapter 3, Vapor Pressure	P. A. Gupte, T. E. Daubert, R. P. Danner	1982
Chapter 4, Density	N. O. Umesi, R. P. Danner, T. E. Daubert	1982
Chapter 5, Thermal Properties	P. A. Gupte, T. E. Daubert, R. P. Danner	1983
Chapter 8, Viscosity	V. Bhethanabotla, T. E. Daubert, R. P. Danner	1985
Chapter 9, Thermal Conductivity	M. Nagvekar, T. E. Daubert, R. P. Danner	1985
Chapter 10, Diffusivity	D. P. Northup, A. J. Engel, R. P. Danner, T. E. Daubert	1986
Chapter 7, Surface Tension	M. S. High, M. Nagvekar, R. P. Danner, T. E. Daubert	1987

[a] Monographs documenting the selection of the contents of the *API Technical Data Book - Petroleum Refining* are available from University Microfilms, Ann Arbor, MI. [b] Monographs documenting the basis for the selection of the contents of the *Manual for Predicting Chemical Process Design Data*, Design Institute for Physical Property Data, American Institute of Chemical Engineers, New York, NY.

If critical properties as well as vapor pressures and liquid densities over the entire applicable temperature range were measured for a large group of multifunctional organics and higher molecular weight single function organics, methods for property prediction could be improved substantially. Although heat of vaporization can be exactly calculated from the Clapeyron equation from vapor pressure and densities, additional wide range data on enthalpy of vaporization would be desirable to validate prediction methods proposed. Emphasis should be placed on obtaining such pure component data over the entire range of temperature from the triple point to the critical point.

While improvements in prediction methods are necessary for pure compounds and defined mixtures for almost all properties, a single correlational framework for pure component properties, especially liquids, is desirable and would aid a great deal in adapting the predictive schemes for simulators. As pure compound predictions are the primary emphasis of this paper, review of the available methods shows some theoretical developments for vapor property predictions. Primarily empirical methods have been developed for liquid property predictions where molecular interactions yet defy good theoretical treatment. Second-order group contribution methods have been very successful at least for (1) ideal gas heat capacity, entropy, and enthalpy of formation, (2) second virial coefficient, (3) critical properties, (4) liquid viscosity, and (5) liquid thermal conductivity. The extension of such methods to (6) vapor pressure, (7) liquid heat capacity, and (8) liquid density as well as the continued upgrading of the existing methods to compounds with several functional groups and of higher molecular weight would be a great service to prediction of all physical properties. The necessity that certain thermodynamic properties must be highly accurate while most transport properties can be less accurate must be considered as such work proceeds.

Once a single consistent correlational framework for estimating pure component properties is developed, standardization of rules for defined mixtures would be a laudable goal. Although current mixing rules for defined mixtures are reasonable, no consistency whatsoever exists even for similar properties.

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