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Uncertainty Reporting for Experimental Thermodynamic Properties[†]

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This paper reviews practices in the expression of uncertainty in the experimental literature for thermodynamic property measurements with determinations of critical temperature for pure compounds used as a case study. The time period considered is from 1940 to the present, with an emphasis on results published since 1990. One hundred ninety-four articles were considered involving hundreds of compounds. The goals of this paper are to show how the nature and extent of estimations of uncertainty have changed in this time period and to document the extent to which the information provided in the articles corresponds to the recommendations of the GUM. (The GUM is the *Guide to the Expression of Uncertainty in Measurement* published in 1993 by the International Organization for Standardization, ISO. This document describes current recommended practices in this area.) It is shown that although gradual and continuous progress has been made in the reporting of uncertainty information, comprehensive uncertainty analyses remain rare, particularly with regard to the consideration of contributions arising from sample impurities. Problems in the application of experimental property results to technological challenges are discussed within the broad context of inconsistencies in the expression of uncertainty. Widespread acceptance and application of the standard methods and terminologies established within the GUM are encouraged for the benefit of experimentalists, data evaluators, and data users alike.

Introduction

Estimates of uncertainty are the measure of data quality for all experimentally determined quantities and form the basis for the understanding, evaluation, and application of all scientific data. With adequate knowledge of uncertainty, comparison and evaluation of results from different laboratories can be realized, from which a fair judgment may be achieved concerning the level of confidence in the property values. Only then, with appropriate application of statistical methods for the propagation of uncertainties, can the results be applied to the solution of technical problems. Although few scientists would disagree that the reporting of reliable uncertainty information is important, it is broadly recognized that the nature and extent of that reporting in the literature is highly variable. This recognition is most apparent to those involved in the critical evaluation of property data, whereas it is often less clear for those involved in property-data applications, such as designers of chemical processes and environmental policy analysts, including those attempting to create science-based policies for key issues such as environmental regulations, ecosystem restoration, gasoline formulation, control of greenhouse gases, and plant and automotive emission standards.

The quality of predictive models and chemical process designs depends on the reliability (i.e., the uncertainties) of the physicochemical property data that support them. Whiting and colleagues[1] have reported a series of studies concerning uncertainty analyses of experimental data, predictive models, and process simulations that demonstrate the importance of the careful consideration of uncertainties. Recently, in a review of industrial needs in the area of physical properties, Gupta and Olson[2] pointed out that "Increased emphasis is being placed on accurate physical property data and models due to increasing capability and complexity of chemical process simulation software, trends in process simulation applications, and nontraditional applications of physical properties." Underlying any such discussions involving uncertainty and accuracy is the assumption that all parties have a common understanding of what is meant by these terms. It will be shown that this assumption is difficult to justify on the basis of the published literature.

Periodically, those involved in the application of property data express surprise when confronted with inconsistencies in reported property values. Typically, however, little results from these observations because the community of data producers (experimentalists) and data users (process design engineers and analysts) are only weakly linked. This was evidenced recently in reactions[3] to a report[4] related to the reliability of experimental property data used as a basis for predicting environmental impact and fate. The report[4] reviewed all known aqueous solubilities and octanol−water partition coefficients for the pesticide DDT and its persistent metabolite DDE. The report concluded that "The accuracy and reliability of the vast majority of the data are unknown due to inadequate documentation of the methods of determination used by the authors."

Predictably, in reaction to this report, two viewpoints were expressed:[3] (1) those who claimed that inconsistencies were always a problem but could be "worked around" and (2) those who believed that the poor quality of the data

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cast doubt on the scientific validity of the "workarounds" and, consequently, invalidated any analysis. Both groups made the further predictable request for more and higher-quality data. None of these proposed solutions is promising, and the entire scenario could have been avoided if reliable uncertainties had been provided with the original experimental reports. Contributors to the *Journal of Chemical and Engineering Data* (and other journals of similar scope), as experimentalists, are in the best position to improve this unfortunate and common situation.

The general concept of uncertainties has been recognized and discussed by many generations of scientists, but the establishment of any broadly accepted standards for their formulation or even terminology has been very difficult to achieve. The annotated bibliography compiled by Cali and Marsh[5] and published in 1983 includes numerous citations to discussions of uncertainty representation in the literature. In 1978, an international committee was formed, and after years of negotiation and discussion among national measurement institutes and related scientific bodies worldwide, the *Guide to the Expression of Uncertainty in Measurement* (commonly referred to as the GUM)[6] was published in 1993, together with the *International Vocabulary of Basic and General Terms in Metrology*,[7] by ISO (the International Organization for Standardization). These ISO recommendations were adopted with minor editorial changes as the *U.S. Guide to the Expression of Uncertainty in Measurement*.[8] The recommendations have been summarized in *Guidelines for the Evaluation and Expression of Uncertainty in NIST Measurement Results*,[9] which is available for free download from the Internet (http://physics.nist.gov/cuu/).

Recently, we summarized the recommendations of the GUM with particular application to the reporting of experimental thermodynamic property data.[10] This was the second in a series of papers that established XML (i.e., extensible markup language) representations (ThermoML) for the storage and exchange of thermodynamic property data[11] together with uncertainty information based on the GUM recommendations. The third paper in this series concerning the representation of predicted and critically evaluated thermodynamic data, including equation representations, has now been published.[12] ThermoML was recently accepted as the basis for an IUPAC standard for the storage and exchange of thermodynamic property data.[13]

To develop well-defined data structures within ThermoML for the representation of uncertainty information, it was necessary to interpret the formulations of the GUM with specific application to thermodynamic property measurements.[10] The reader is referred to ref 10 for full descriptions of the terms used in the present paper. See *Basic Principles and Definitions* given on pages 1345 to 1348 of the paper.[10]

The most comprehensive expressions of uncertainty are the combined standard uncertainty u_x and the combined expanded uncertainty U_x. u_x can be represented as the mathematical expression

$$u_x = f(x_1, x_2, x_3, ...) \qquad (1)$$

where the symbols x_i represent various contributions to the uncertainty that are appropriately weighted to estimate u_x. For example, the estimated uncertainty for a temperature value might be a function of the method and traceability of the sensor calibration, the instrument used to read its response, estimated gradients in the apparatus, effects of thermal inertia, and so forth. In the case of a measured critical temperature, this quantity must also include uncertainties associated with the quality (i.e., the purity and thermal stability) of the sample employed. A well-designed experiment (i.e., one that includes the identification and control of the largest contributions x_i in eq 1 through the determination of values of $\partial u / \partial x_i$) will reduce uncertainties and will improve the quality of the uncertainty estimates, but some scientific judgment is always involved in estimating u_x.

u_x represents 1 standard deviation and is related to U_x through the expression

$$U_x = u_x k_x \qquad (2)$$

The coverage factor k_x is a numerical multiplier used to expand u_x with a specified level of confidence (usually 95%), which is an estimate of the probability that the measurand is within a specified range. The measurand is sometimes referred to as the "true value", the exact value of which is unknowable by definition because of the very existence of finite (nonzero) uncertainties in all attempts to measure the value.

Two additional quantities are relevant to the discussion in the present paper, and these are device specifications and repeatabilities. Device specifications are statements of uncertainty for a particular device, such as a thermometer or pressure gauge, provided as part of a calibration, typically from the device manufacturer. Repeatability is defined as the "Closeness of the agreement between the results of successive measurements of the same measurand carried out under the *same* conditions of measurement."[7] (The italics were added here for emphasis.) It is imperative to recognize that these quantities represent types of precision and are, therefore, only two of many components that might be considered in the evaluation of the comprehensive combined expanded uncertainty.

As part of our work in the critical evaluation of thermodynamic property data within the Thermodynamics Research Center (TRC) at NIST, we estimate the combined expanded uncertainty for experimental property values reported in the literature based upon the metadata provided by authors within articles, including descriptions of the samples used and accumulated knowledge of the experimental methods applied. These uncertainty estimates are often significantly larger than the uncertainties, accuracies, or precisions commonly reported in articles. In the present paper, we will show that the source of this apparent discrepancy lies in the very common practice of incomplete reporting of uncertainty information together with inconsistent use of the vocabulary related to uncertainty representation.

It should be noted that the terms *precision* and *accuracy* are not recognized as quantitative measures of uncertainty within the GUM.[6] The term precision is not used within the GUM because of its numerous and conflicting meanings, whereas the term accuracy is defined as the "closeness of agreement between the result of a measurement and the measurand" and is, by definition, unknowable because the true value of any measurand is unknowable.

The present paper reviews practices in the expression of uncertainty in the experimental literature for thermodynamic property measurements with determinations of critical temperature for pure compounds used as a case study. The time period considered is from 1940 to the present with emphasis on results published since 1990. Goals of the paper are to show how the nature and extent of estimations of uncertainty have changed in this time period and to document the extent to which the information provided in the articles corresponds with the recommendations of the GUM. A broader goal is to encourage more widespread acceptance and application of the standard methods and terminologies established by the GUM.

We wish to make clear that the purpose of this paper is not to criticize authors for what were and are commonly accepted practices in the reporting of uncertainty information. The publication of the GUM in 1993 was the result of a project that was begun formally in 1978, which implies that the need for such a consensus document was recognized well before even this date. It is true that noncomprehensive reporting practices for uncertainties (e.g., reporting of repeatabilities only or failure to consider all major uncertainty components) were and continue to be considered acceptable by many researchers and journals, but this contradicts the fact that this limited information is not adequate for modern technological applications.

Scope of the Literature Review

The measurement of critical temperature T_c was chosen as the subject of this case study for three reasons. First, T_c is one of the most fundamental properties in thermodynamics and is commonly used in corresponding-states correlations for the prediction of numerous properties of industrial interest. Consequently, the uncertainty in T_c not only represents the data quality for T_c itself but also has a significant effect on the quality of derived property data and models through propagation. Second, there have been a relatively small number of reports of experimental T_c measurements since Cagniard de la Tour first observed the phenomenon of the critical point in 1822[14] (approximately 600 articles involving 2000 measurements for a total of 650 compounds), which makes it possible to cover a large fraction of the literature in the present study. Finally, a large number of thorough reviews[15] on T_c have been published in the period 1923 through 2001. Many of these reviews included critical reviews of progress in measurement technology as well as the reliability of experimental results. References 15f−m represent the results of an ongoing IUPAC project in this field, with three additional articles planned.[16]

Articles published since 1940 for a wide variety of pure compounds were chosen for consideration within the present project. A total of 194 articles were considered, which corresponds to approximately one-third of the entire published literature for experimental critical temperature determination and represents approximately 40% of those published since 1940. The complete list of chosen references is included in the Supporting Information provided with this article. Emphasis was placed on articles published in the 1990s and 2000s (73 articles) to have a particularly good representation of recent practices.

Method of Information Collection and Categorization

Each article was reviewed carefully for uncertainty information, and the type of information provided was categorized into five groups. These five groups were ranked on the basis of the extent to which the reported information corresponds to the modern recommendations of the GUM. The five categories are (in order of least-complete to most-complete information) as follows:

(1) No info: No uncertainty information of any kind was reported. In these papers, only a numerical value for T_c was given.

(2) Device specification: Only calibration information for the temperature-measuring system was provided or discussed.

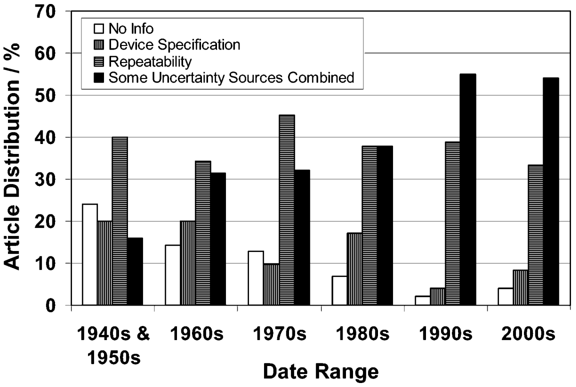

Figure 1. Article distribution over four categories of uncertainty reporting. The categories are defined in the text. The fifth category (complete combined uncertainty) is not represented because only one article was ranked in this category.

(3) Repeatability: Repeatability (a type of precision) was the most extensive uncertainty information provided.

(4) Some combined uncertainty: Uncertainty components from multiple sources were considered, but the analysis was incomplete. The most common omission in this category was a consideration of the quality of the chemical sample.

(5) Complete combined uncertainty: A comprehensive consideration of uncertainty sources was provided, including explicit consideration of the chemical sample.

The rank of a particular paper was based on the highest level of information provided. For example, if a device specification (rank 2) was provided together with the repeatability (rank 3), the paper was ranked 3. Articles that included comparisons with critically evaluated values from the literature to establish the effectiveness of the method were generally given a rank of 4. This type of validation is explicitly described in the GUM as a type-B evaluation.

Assignment of rank for many articles necessarily involved considerable subjective judgment. The meanings of phrases such as "temperatures were measured to...", "the absolute accuracy was...", "temperatures were controlled to within...", and so forth are not clear and could readily imply a device specification, a repeatability, or some kind of partial combined uncertainty. Uncertainty discussions in many articles involving visual techniques were comprised exclusively of a discussion of the appearance and disappearance of the meniscus. Reported uncertainties in these cases were assumed to be repeatabilities and were ranked 3. In all cases, the context of the discussion was taken into account to make a best guess concerning the meaning of the information provided.

Results

Results are shown graphically in Figures 1 and 2 and were grouped on the basis of the decade in which they were reported. Reports from the 1940s and 1950s were combined. The total number of articles considered in each time period is given in Table 1. Figures 1 and 2 provide similar information. In Figure 2, articles ranked 2 (device specification) and 3 (repeatability) were combined into a single category (precision). Device specification and repeatability are both indications of precision, and the combination of these categories made certain trends more apparent. Of all articles considered, only one[17] was ranked 5 (complete combined uncertainty). Consequently, this category is not represented in the Figures because the bar in the graph would not be visible.

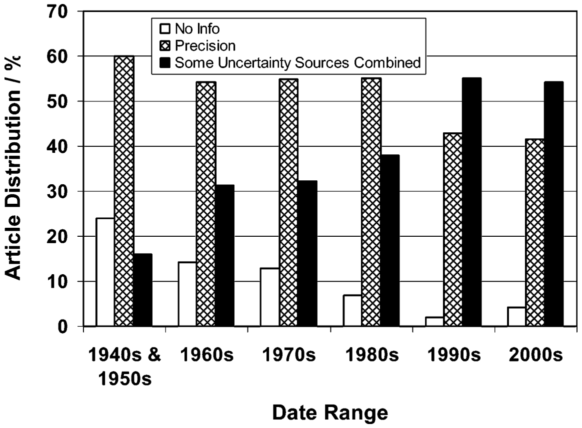

Figure 2. Article distribution for three categories of uncertainty reporting. Relative to Figure 1, device specification and repeatability were combined into the single category precision. The categories are defined in the text.

Table 1. Time Distribution of Articles Considered in This Case Study

time period	number of articles
1940s and 1950s	25
1960s	35
1970s	32
1980s	29
1990s	49
2000s	24
total	194

Discussion

Several positive trends are apparent in both Figures. The percentage of articles providing no uncertainty information (rank 1) has dropped dramatically from approximately 25% in the 1940s and 1950s to near 3% since 1990. Similarly, the percentage of papers reporting some type of combined uncertainty (rank 4) has increased from near 15% to approximately 55% in the same time period. It is also apparent that although the reporting of uncertainty information has improved with each decade there is still much room for improvement. For example, in the time period since 1990, approximately 42% of the published articles report only some type of precision information (ranks 2 and 3), as shown in Figure 2. This restricted information on precision provides only a lower bound for the combined standard uncertainty and is of limited value to data evaluators and application engineers. Unfortunately, this fact is not often recognized by data users, which results in the sort of ambiguities described in the Introduction concerning octanol−water partition coefficients.[3,4]

An important finding of this case study is the nearly complete absence of consideration of sample quality in the discussions of uncertainty. In some cases, the authors described extensive purification procedures together with careful analyses to demonstrate that their samples were of very high purity, sometimes as high as 99.99 mol %. In such cases, the absence of an explicit discussion of the contribution of sample impurities to the combined uncertainty is understandable but still cannot be completely ignored. Some claimed uncertainties were on the order of a few millikelvins, for which the effects of even such low levels of impurity can be important. Conversely, it remains common for authors to report that their sample was a commercial product of some purity stated by the manufacturer (sometimes as low as 96 or 97%) and that it was used without further purification or confirmation of the manufacturers claim, while listing “uncertainties” on the order of 0.1 K or even less. There are numerous reports concerning critical temperatures for mixtures in the literature[18] that can be used to show that an impurity of even 0.1 mol % can change the measured critical temperature by 0.1 K.

A final observation is that the recommendations expressed in the GUM[6] and the *International Vocabulary of Basic and General Terms in Metrology*[7] have been generally ignored since their publication in 1993 in the reporting of experimental critical temperature determinations. Although the topic of uncertainties has gradually been given more emphasis with time, it is clear that most longstanding problems associated with clear communications of the meaning of reported uncertainties remain.

It is our hope that this case study will encourage experimentalists, reviewers, editors, data evaluators, and data users to consider carefully the application of the international recommendations for the expression of uncertainty[6,8] and vocabulary[7] that have taken many years to develop. The purpose of communication standards is not to burden those involved in the reporting and estimation of uncertainties but to increase the value of their efforts greatly through clear and unambiguous definitions and to prevent the reduction in value of their results that accompanies the absence of information about uncertainty. We hope that it is clear from the discussion presented in the Introduction that these issues extend far beyond simple comparisons of multiple results for a given quantity. If thermodynamic property results are to be used with confidence to help solve the important technical problems facing society today (environmental quality, control of greenhouse gases, plant and automotive emission standards, transportation fuel formulation, etc.), then it is clear that unambiguous uncertainty expressions are essential. In the report that initiated the discussion concerning the quality of solubility and octanol−water partition coefficient data,[4] Pontolillo and Eganhouse recommended that much greater efforts are needed to establish and maintain consistent reporting requirements for physicochemical property data. With broad international agreement now established concerning methods for the expression of uncertainty (the GUM)[6,8] and the associated vocabulary,[7] the means to establish and maintain consistent reporting requirements for physicochemical property data are in place. The next step is to apply them universally.

It is important to emphasize that only comprehensive formulations of uncertainties (complete combined uncertainties) provide the full measure of data quality. If these are propagated into uncertainties for properties related to industrial streams, then this can lead to enormous economic benefits in the implementation of results of chemical process simulations, particularly for optimal equipment selection. The implementation of this possibility can fundamentally change the nature of future chemical process modeling and design.

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Supporting Information Available:

The complete list of reports of experimental critical temperature determinations used in the compilation of the statistics in this paper is available. This material is available free of charge via the Internet at http://pubs.acs.org.

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