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Quality assurance in thermodynamic databases for performance assessment studies in waste disposal*

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Abstract: Performance assessment studies in underground disposal of radioactive or toxic waste need to consider all reactive interactions between waste and its surroundings. Thermodynamic equilibrium and reaction path calculations represent an important tool for this purpose. The reliability of the results depends first of all on the quality of the thermodynamic database used for the calculations. Several quality criteria of thermodynamic databases are discussed in connection with the characteristics of current database projects [Nuclear Energy Agency Thermochemical Database (NEA-TDB), Yucca Mountain database, Dortmund Databank (DDB), Common Thermodynamic Database (CTD), FreeGS, and Thermodynamic Reference Database (THEREDA)] including the situation for molten salts. The future role of the IUPAC standard for thermophysical and thermochemical data storage is emphasized.

Keywords: thermodynamic databases; radioactive waste disposal; geochemical databases; ThermoML; performance assessment studies.

INTRODUCTION

Isolation of radioactive and toxic waste from the biosphere for a very long time represents a challenge for all industrial developed nations. Most concepts are based on underground disposal of the waste in repositories in deep geological formations. In assessment studies, alternative concepts have to be eval-

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uated in terms of safety and economy. Owing to the different geological prerequisites in a country's geology, a variety of different geological environments have to be considered for disposal: rock salt formations, granitic formations, or clays. Reactive interaction of the various wastes and its compartment materials with the complex composed geological environment have to be predicted over thousands of years. The most developed tool for such predictions represents the chemical thermodynamics with reaction modeling based on this concept. Modern numerical codes have no principal problems in solving the respective equations for calculation of the reaction and phase equilibria even in the presence of hundreds of chemical species and transport along a concentration gradient. The reliability and quality of the results depends first of all on the quality of the thermodynamic database underlying the calculations. Since many mineral phases and aqueous solution species from most of the elements of the periodic table are involved in these interactions, extended thermodynamic databases of high reliability are required.

Thus, databases have to be developed to meet the following requirements:

- correct description of simple systems (2–4 components)
- description of multicomponent and multiphase systems
- containment of all necessary standard data of mineral phases and aqueous species
- containment of interaction coefficients for all aqueous phase species for an appropriate model
- compatibility with existing calculation codes
- quality assignments to the data
- defined updating procedures for incorporation of new data
- easy access for users
- traceability to the data sources

In this paper, the present status of thermodynamic databases as discussed at a workshop is reviewed and efforts for further developments are highlighted. In order to learn as much as possible from different approaches, not only geochemical-oriented databases are analyzed.

TYPES OF THERMODYNAMIC DATABASES

In principle, all measured and derived thermodynamic or equilibrium properties can be the subject of a systematic collection, which then represents itself as a database. Depending on the type of data, one can distinguish between databases of

A original experimental data;
B evaluated experimental data, recommended data;
C thermodynamic standard data of chemical species; or
D model parameters for thermodynamic excess quantities

For instance, the IUPAC-NIST "Solubility Data Series", Vols. 1–85 [1] represents a combination of type A and B databases. In these books, all available solubility data for the systems under consideration are compiled and evaluated for recommendation of "best" data. Table books such as NEA-TDB [3], CODATA [4], JANAF [5], Barin [6], and Gurvich [7] are examples of type C databases. The use of thermochemical standard data for calculations of phase or reaction equilibria in the presence of mixed liquid phases requires the application of suited species interaction models (Pitzer [8], SIT [9], UNIQUAC [10], UNIFAC [11], and others [12,13]). These model or interaction parameters are evaluated from appropriate thermodynamic measurements in solutions or liquid mixtures. Tabulations of such model coefficients are databases of type D, for instance, Pitzer coefficients [8], SIT coefficients [9], or UNIQUAC parameters [14]. Further criteria such as application fields (chemical process simulations, geochemical simulations, material property prediction), end user categories, or classes of substances govern the database characteristics.

CHARACTERISTICS OF SOME EXISTING DATABASES

OECD Nuclear Energy Agency Thermochemical Database (NEA-TDB) project (<www.nea.fr/html/dbtdb>)

The objective of the NEA-TDB is to make available a

- comprehensive,
- internally consistent,
- internationally recognized, and
- quality-assured

chemical thermodynamic database for selected elements to meet the modeling requirements for safety assessments of radioactive wastes. Another aspect of the efforts is to promote exchange of information on related activities being carried out in OECD NEA Member countries. The project started in 1986 with TDB I and at present has reached stage III. Typically, for the selected elements international review teams are formed, who are producing critical evaluated thermodynamic standard data strictly following the NEA-TDB guidelines [3]. Guidelines exist for

- the review procedure and data selection,
- the extrapolation to zero ionic strength,
- the assignment of uncertainties,
- temperature corrections to thermodynamic data and enthalpy calculations,
- standards and conventions for TDB publications, and
- the independent peer review of TDB reports.

Thermodynamic quantities chosen for the database are reaction data (log K°, $\Delta_r G_m^\circ$, $\Delta_r H_m^\circ$, $\Delta_r S_m^\circ$, $\Delta_r C_{m,p}^\circ$), formation data ($\Delta_f G_m^\circ$, $\Delta_f H_m^\circ$), and the absolute molar entropies and heat capacities (S_m°, $C_{m,p}^\circ$). Procedures of selection and evaluation are described in detail in the corresponding books for the elements. Until now, nine such volumes have been published covering the elements U [15,19], Am [16,19], Tc [17,19], Np, Pu [18,19], Ni [20], Se [21], Zr [22], and one volume with metal complexes with organic ligands [23]. The review teams have to derive the standard data from sources reporting the original data. Normally, the data are accomplished with uncertainties on a 95 % confidence level. All necessary auxiliary data are listed in every volume, thereby as far as possible compatibility with the Commission on Data for Science and Technology (CODATA) is envisaged. For ionic strength extrapolations in aqueous systems, the SIT model is used [9] and the corresponding coefficients are reported.

The project is managed by an "executive group" acting as a scientific advisory board and a "management board" responsible for funding and strategic decisions. For the daily coordination of the review teams acts a project coordinator.

Yucca Mountain project thermodynamic database

For performance assessment of an underground radioactive waste repository in the Yucca Mountains (USA), a database was developed containing all necessary thermodynamic and some kinetic data for performance assessment. This comprises actinides, main inorganic elements, a few organics, and about 2000 minerals related to the geology of the Yucca Mountains. The program to establish the database was partly a continuation of the earlier finished salt repository program (WIPP site, Carlsbad). A large number of institutions and universities were involved in literature search and data evaluation. The database itself represents input files (DATA0.ymp, DATA0.ypf [25], DATA0.ypa) for the geochemical calculation code EQ3/6. The files are not freely accessible to the public. The main database DATA0.ymp is designed for the use of the extended Debye–Hückel equation to correct for ionic interactions in aqueous solutions. The other two files contain data derived for use with Pitzer equations.

Quality checks were performed by comparison of simulated and experimental data for special situations and systems, for example, [24]. No special quality guide was followed in the evaluation procedures, and no effort was made to reach consistency within the database.

Dortmund Databank (DBB) (<www.ddbst.com>)

The Dortmund Databank represents a commercial database. The first activities started in 1973 at the University of Dortmund with the development of a database for group contribution G^E-models (UNIFAC) to calculate phase equilibria in systems of organic compounds for industrial processes such as distillation and extraction. Later on, the model base was extended and software for data handling and representation was developed. With the LIQUAC and LIFAC [26] models, a bridge to the electrolyte systems was built. In 1989, all the activities shifted to the DDBST GmbH in Oldenburg (<www.ddbst.de>) as well as to the Institute of Chemical Technology at the University of Oldenburg. The database stores original data (52 000 references, more than 100 000 data sets) as well as derived thermodynamic data. For pure components, other physical and transport properties are also stored. Figure 1 gives an indication of the databases content. For the users, a series of retrieval and property estimation programs as well as application programs for process synthesis, etc. are supplied. Complex software enables fast incorporation of new experimental data, and it gives consistent derivation of model coefficients for the database. More than 10 people are working full time for the database and software. The further development of estimation methods for real mixture behavior is sponsored by a consortium of more than 45 companies (<www.unifac.org>). For the experimental determination of new data for commercial customers, an additional company was founded (<www.ltp-oldenburg.de>).

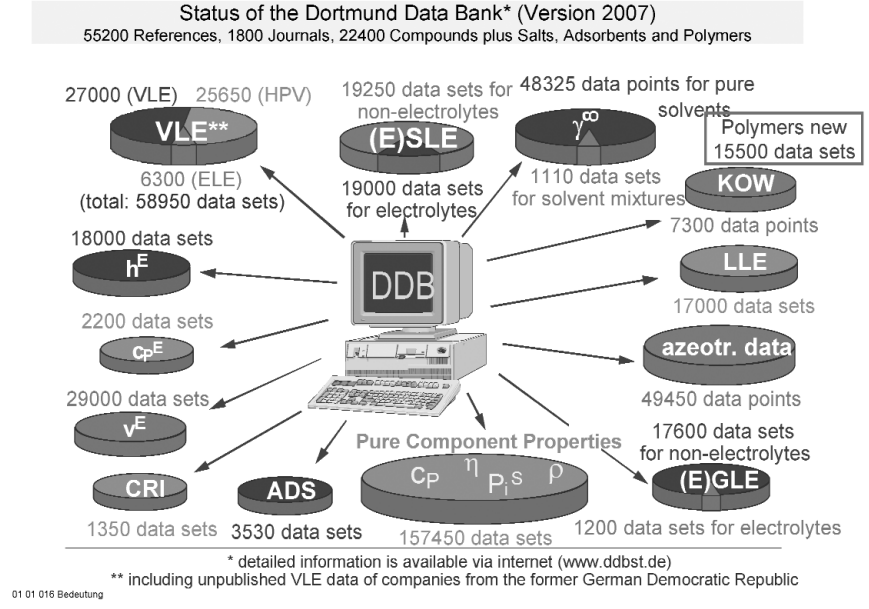

Fig. 1 Content of the Dortmund Databases.

Common Thermodynamic Database (CTD) project (<http://ctdp.ensmp.fr>)

The project was launched in 2003 by J. van der Lee at the Paris School of Mines/Center of Geological Computing (ENSMP/DIG). It officially started in 2005 with five funding partners (ARMINES, CEA, eDF, IRSN, and TOTAL). The main objectives of the project are to

- centralize collective efforts to elaborate a robust TDB for (geo)chemists;
- use a common (Web-based) review platform that allows federate expert groups and users of speciation models to develop the database;
- allow storage of all types of data, including experimental data;
- maintain internal consistency (source-proposal mechanism) and traceability to the original source; and
- be in a model-independent format (XML, see Fig. 2).

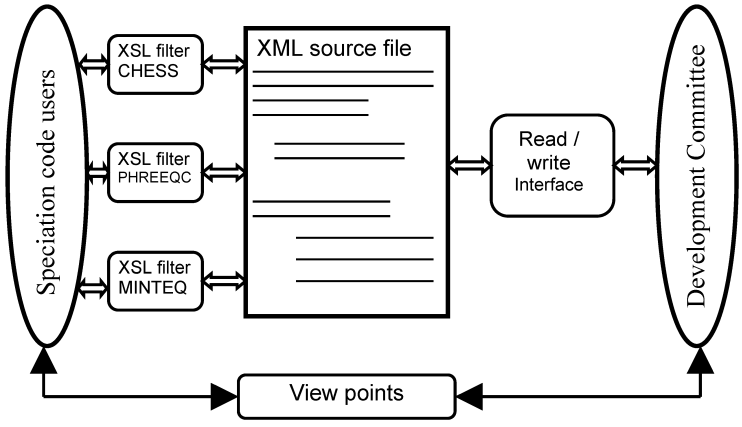

Fig. 2 Data flow sheet of the CTD project.

Seven working groups constitute the organization of the project (Fig. 3). The current content can be characterized as a preliminary one that is about 1500 aqueous species, 1750 organic species, 1500 solids, and 180 gaseous species. Data were taken from other databases such as NEA-TDB, NAGRA/PSI, LLNL (EQ3/6), GEOPIG, and IUPAC. Two categories of access exist: experts and end-user. Experts contribute to the database and have access to the "working database". The end user will get an updated official release every 6–12 months, which can be freely downloaded through the Internet after registration. Several levels of confidence are indicated:

0 new submission by one of the members,
1 acceptance of the submission by the coordinator (but the data is not reviewed yet by the other experts),
2 the data has been reviewed but there is no common consensus (link to the ongoing discussion), and
3 the data has been reviewed and there is common consensus for acceptance.

Four grades of consistency levels are automatically calculated by the system software. Several filters are provided for the user to use the database in various codes (CHESS, PHREEQC, EQ3/6). In order to fill data gaps, the inclusion of estimated data from correlations and analogies is allowed.

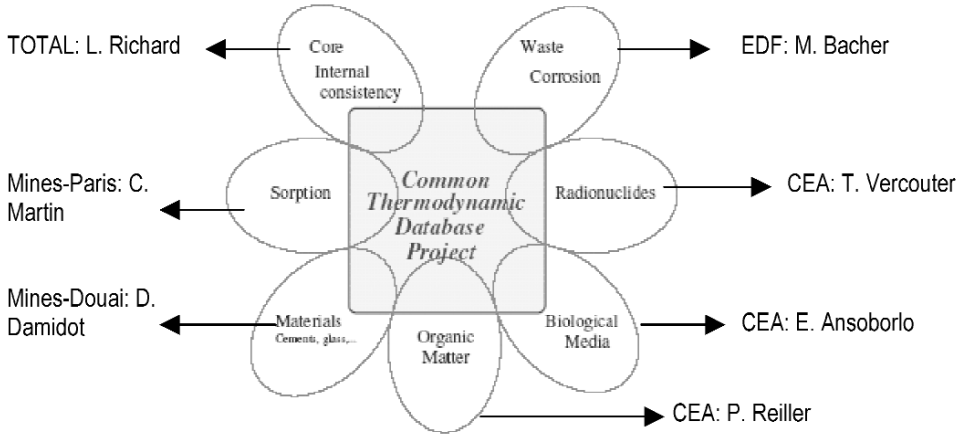

Fig. 3 Organization of the CTD project.

FreeGs thermodynamic database project (<http://www.ga.gov.au/minerals/research/methodology/geofluids/freegs_about.jsp>)

FreeGs represents an Australian initiative under the auspices of "Geoscience Australia", a government organization. Other institutions and universities are involved in the common activity. The FreeGs thermodynamic database project is part of a "virtual centre for geofluids and thermodynamic data" raised by pmd*CRC ("predictive mineral discovery*Central Research Centre"). FreeGs shall provide a database freely accessible through the Internet. The content will cover thermodynamic standard data for all important mineral, gaseous, and aqueous species over a wide range of temperature and pressure. Versions with recommended data and collections of available data are planned. Different thermodynamic models and various types of users (casual, novices, experts) can be served by the database. Thus, FreeGs will be a common platform for the Australian geoscience community (Fig. 4).

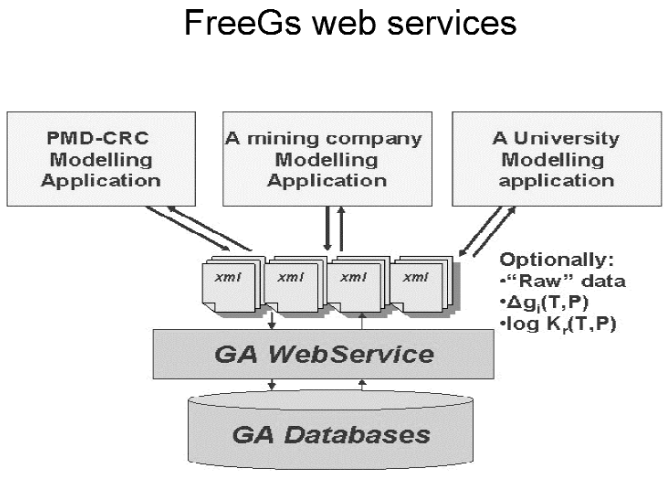

Fig. 4 FreeGs data and service concept.

FreeGs is NOT a self-consistent database of thermodynamic properties, nor is internal consistency a goal of the project, though they will strive for a "reasonable" consistency wherever possible.

The values, which will be recommended for the pmd*CRC community, are "reasonable" from a modeler's point of view (i.e., they reproduce the natural or experimental equilibria to the satisfaction of the maintenance team). For external users, the choice of the selected data sets and responsibility for this choice is entirely with the user.

The constants and values in FreeGs are not forcibly certified, instead a confidence level is attributed.

At present, a "FreeGs thermodynamic calculator" is accessible on a Web site. Registrated users can access the database forms (Oracle-based), search for data sets, and add their own. Web services for specific user tasks are under development (e.g., the delivery of automatically generated sets of independent chemical reactions to describe the equilibria in a given chemical system).

Molten salt databases

Database work in the field of molten salts had been started about 1975 by Janz at the Rensselaer Polytechnic Institute in Troy, NY (USA). In connection with a program for selection of standard data for certain properties such as electrical conductivity and solid–liquid phase diagrams, table books were published, which later were transferred to electronic medium by National Institute of Standards and Technology (NIST). A consistent thermochemical database was only developed by Pelton et al. [27] for the multicomponent systems LiCl, NaCl, KCl, $MgCl_2$, $CaCl_2$, $MnCl_2$, $FeCl_2$, $CoCl_2$, and $NiCl_2$. This database is distributed commercially in combination with the thermochemical calculation program FactSage [28].

THEREDA—the German database project for performance assessment studies

The project THEREDA (Thermodynamic Reference Database) is planned and supported by the following major research institutions working in the field of radioactive and toxic waste disposal in Germany:

- Gesellschaft für Reaktorsicherheit, Institut für Tieflagerung Braunschweig (GRS-B)
- Forschungszentrum Karlsruhe—Institut für Nukleare Entsorgung (FZK-INE)
- Technische Universität Bergakademie Freiberg (TU BAF)

Initiated four years ago, the concept was worked out during regular meetings of the project partners. The German Federal Ministry of Labour, the Federal Ministry of Education and Research, and the Federal Ministry of Environmental Protection and Reactor Safety have been involved in the preparation of the project in an early stage. The project has been financed by these authorities since 2006.

The main objective of THEREDA is the generation of a comprehensive and internally consistent thermodynamic reference database for geochemical modeling of near- and far-field processes occurring in repositories for radioactive wastes in different rock formations currently under discussion in Germany. Special emphasis will be laid upon a complete documentation and traceability of the registered data. It is planned to integrate data from existing qualified thermodynamic databases such as the NEA or the PSI-NAGRA database. These data will be completed by the project partners, especially with data for modeling in saline environments. A strategy shall be developed which enables the user to trace back each single data in the parameter files to the primary literature and the respective data evaluation sheet. A group of experts will maintain and update the database far beyond the time span of its generation.

THEREDA will

- enable the prediction of the activity, speciation, and solubility of radionuclides, heavy metals, and other toxic elements to be stored in underground disposal sites;
- be applicable from low to high ionic strength;

- fulfill quality requirements established by the project partners; and
- be the reference database for performance assessment studies in Germany.

Necessary decisions concerning data management and quality management are taken unanimously by the project partners. For the data that will be incorporated into the database, the responsibility lies with the institution that generates these data, as given in the scheme of Fig. 5.

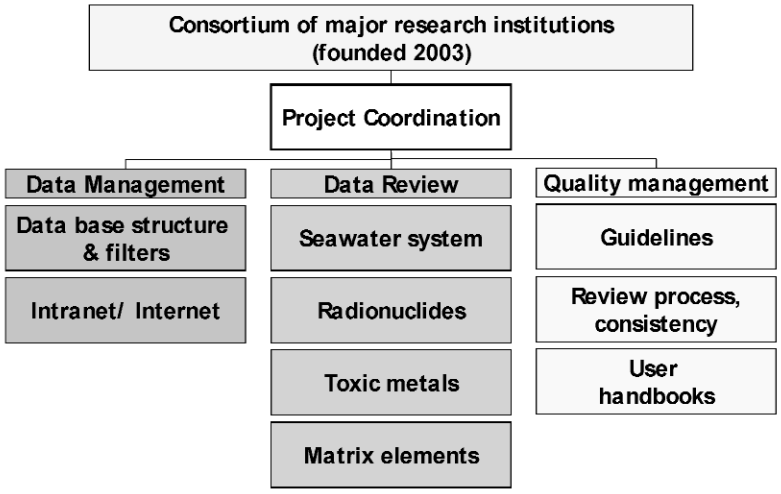

Fig. 5 Organizational scheme of the THEREDA project.

The major constituents H^+-Na^+-K^+-Mg^{2+}-Ca^{2+}-Cl^--SO_4^{2-}-CO_3^{2-}-OH^--H_2O of the sea-water system will be treated. A temperature range between 0–110 °C will be covered. In certain subsystems, the range will be extended to 200 or 250 °C and pressure effects will be included. Emphasis will be put on correct representations of the solid–liquid equilibria. The radionuclides comprise the actinides Th, U, Np, Pu, Am, Cm, Pa, and the fission products I, Se, Cs, Rb, Sr, Sm, Tc, and Ra. The list of toxic metals consists of Cr, Co, Ni, Cu, Zn, Cd, Hg, Pb, and As. Both the radionuclides and toxic metal database will concentrate on ambient temperatures with certain extensions to enhanced temperatures. Finally, the matrix elements are Al, Si, and Fe, including simple aqueous ions and complexes as well as the complicated cement phases.

Since the database must be applicable for low as well as for high ionic strength solutions, SIT and Pitzer ion interaction parameters shall be included.

During the project, tools will be developed which will automatically generate ready-to-use parameter files for important geochemical codes such as EQ3/6, PHREEQC, and Geochemist's Workbench. The parameter files will be available for free download at the project's Web site. In this way, it is intended to obtain useful feedback from users from around the world.

A data review process is the core of the project's quality system. It will be based on quality guidelines the project partners have previously agreed upon. In many aspects, they are identical with the NEA-TDB guidelines (e.g., for the procedures for extrapolation of data to the reference temperature or to zero ionic strength). Contrary to other databases, data gaps in THEREDA will be filled by estimated data, if they are needed for performance assessment calculations. Data of a low quality level will not be excluded, but their quality will be carefully documented. The consistency of THEREDA with CODATA and NEA databases will be ensured as much as possible.

The most important users of THEREDA will be the participants in the project as they work in the major German institutions involved in radioactive waste management. But THEREDA will be a com-

mon and freely accessible database for *all* parties involved in the process of repository licensing. Public access is guaranteed.

ROLE OF ThermoML

ThermoML is an XML-based IUPAC standard for storage and exchange of experimental [29–31] and critically evaluated [32] thermophysical and thermochemical data. ThermoML stands at the current end of a series of efforts by TRC (Thermodynamics Research Center), CODATA (<www.codata.org>), IUPAC, and NIST (<www.nist.gov>) to define a standard format for physicochemical data storage and exchange. An important link in this chain of developments was ELDATA, the *International Electronic Journal of Physico-Chemical Data*, initiated and edited by Henry Kehiaian. However, only the XML-based technology allowed software limitations to be overcome in respect to readability by humans and machine on one side and the ability to expand the definition tree to future necessities on the other side.

ThermoML provides a uniform, well-defined set of data structures (formats) for representation of thermodynamic property data (see Fig. 6). Uniform data structures dramatically ease communication of information, particularly for machine-to-machine communications, and allow simple and direct transfers of thermodynamic property information between all applications using ThermoML. Use of ThermoML can be seen as a major step toward a computerized system for "dynamic data evaluation", which will allow databases to be updated in a more efficient and objective way.

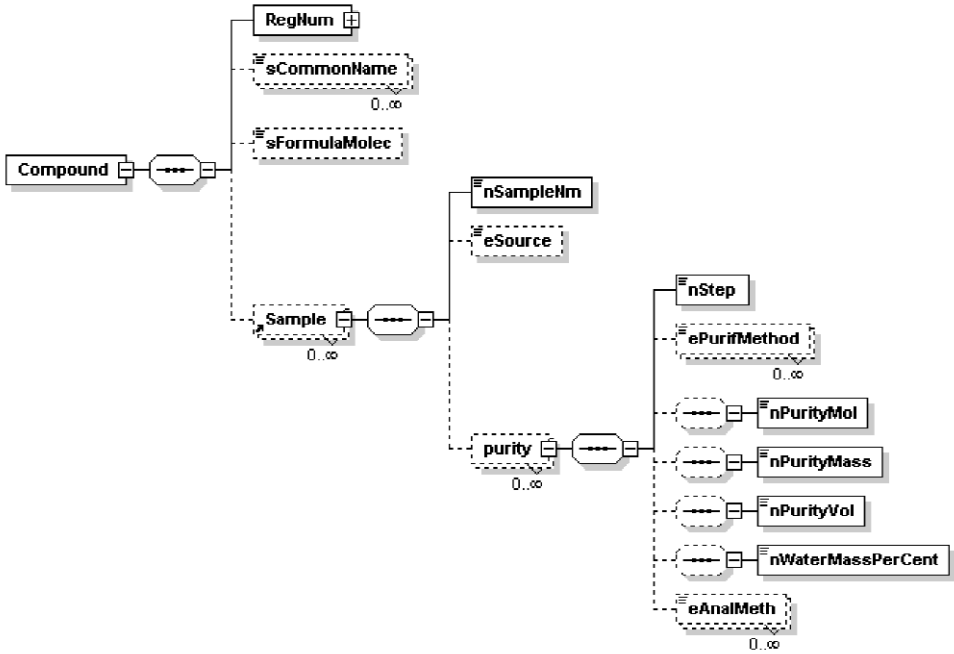

Fig. 6 Compound and sample block as example for meta-data structure in ThermoML.

A detailed description of ThermoML is given in [29–32]. Figure 7 illustrates the application of ThermoML in the communication between authors for publication of data, the journals publisher and data centers. A guided data capture software (GDC, freely downloadable) [33] ensures easy input of all necessary information in the form of a meta-data tree. First-quality checks are performed already at this stage of data storage. Important journals publishing data agreed already to use the system (*Journal of*

Chemical Engineering Data, *Journal of Chemical Thermodynamics*, *Fluid Phase Equilibria*, *International Journal of Thermophysics*, and *Thermochimica Acta*).

At present, ThermoML covers essentially all experimentally determined thermodynamic and transport data for pure compounds, multicomponent mixtures, and chemical reactions. The primary focus at present is on molecular compounds. Polymers and ionic systems including salt and acid solutions are not fully supported at present.

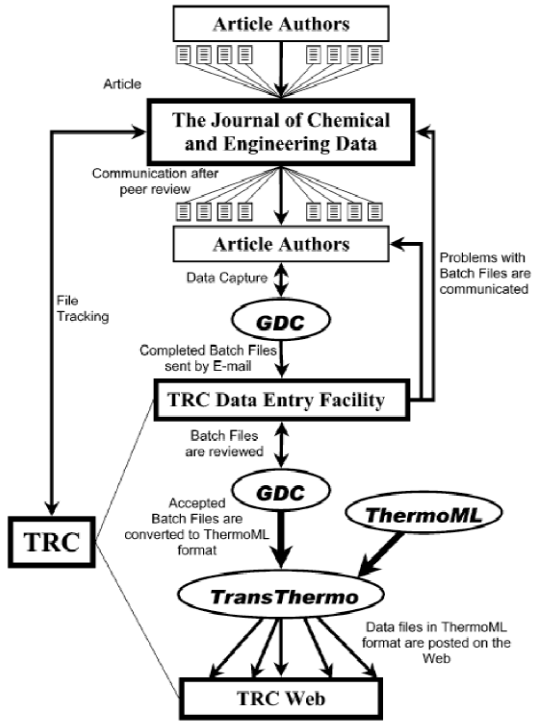

Fig. 7 Information flow between article authors, the *Journal of Chemical and Engineering Data*, and NIST/TRC for data submission and dissemination.

CONCLUSIONS

The status of a variety of actual thermodynamic databases has been discussed. In contemporary database projects, the aspects of

- quality assurance,
- accessibility for users,
- data consistency,
- traceability to the sources,
- completeness of data, and
- updating and maintenance

are treated in different ways. Whereas in the NEA-TDB projects, scientifically best-reasoned standard data are derived from published thermodynamic measurements, in the DDB, standard data and model parameters are stored together with huge amounts of original measured data. NEA convinces the user by the scientific procedure (detail described in books), DDB demonstrates the results of application in a variety of chemical systems as a measure of DDB's quality. The Yucca Mountain's database strategy has to be seen somewhere between these extremes.

NEA-TDB tries whenever possible to assign uncertainties to the data. FreeGs and CTD projects have no special guidelines in this respect. Remarks on confidence levels shall give some impression of the reliability to the user. THEREDA tries to follow the NEA strategy to assign uncertainties and, in addition, to classify the data into definite quality ranges. FreeGs and CTD projects emphasize the fast and easy access to data. The THEREDA project attempts to realize both—easy and free access as well as data quality. Besides uncertainty, data consistency represents another quality feature. In the strict thermodynamic sense, consistency means that data application in different thermochemical cycles yields identical results within the limits of uncertainty. Modern databases should implement such tests as automatic procedures. With the increasing needs and abilities of end users to apply mixture models for calculations with strongly nonideal liquid mixtures, concentrated aqueous solution or solid solution parts of the databases become model-dependent. Consistency in the cases means that the data have to be applied within the same speciation scheme as was used to derive the standard data and model parameters. In the past, databases simply related only to one model. Modern database projects serve several models (DDB, FreeGs, THEREDA). Since mixture and solution models and the respective parameters have specific validity ranges for composition, temperature, and pressure, the database front end should make this transparent and understandable for the users of different professional background also without reading complex manuals.

Updating and maintenance of databases remains the most difficult problem. This activity requires considerable funding. The availability of original measured data in a standardized computer-readable format would be a big jump forward. The IUPAC initiative of ThermoML and the first data-oriented journals using it provide a first basis for this requirement.

From the discussion, it also became clear that the strategy to handle the different quality aspects of a database depends largely on the purpose and the type of clients (users) of the database. There is no "golden way" for all types of material or chemical systems, properties, and users. Performance assessment of waste disposals in the geologic environment requires new features and new challenges. The databases must contain a large number of species, most inorganic and some organic. Constraints for chemical constituents and conditions depend on national disposal concepts and their planning. The time scale reaches from days to thousands of years. Extremely slow equilibration reactions can mostly not be proved in the laboratory. Application of the data in simulations of performance of disposal technologies will identify new critical data, which have to be determined or improved for necessary technological decisions.

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