Thermodynamic Reference Database

Gesellschaft für Anlagen- und Reaktorsicherheit (GRS)

TU Bergakademie Freiberg, Institut für Anorganische Chemie

Helmholtz-Zentrum Dresden-Rossendorf, Institute for Resource Ecology (HZDR-IRE)

Karlsruhe Institute of Technology, Institut für Nukleare Entsorgung (KIT-INE)

AF-Consult Switzerland Ltd.

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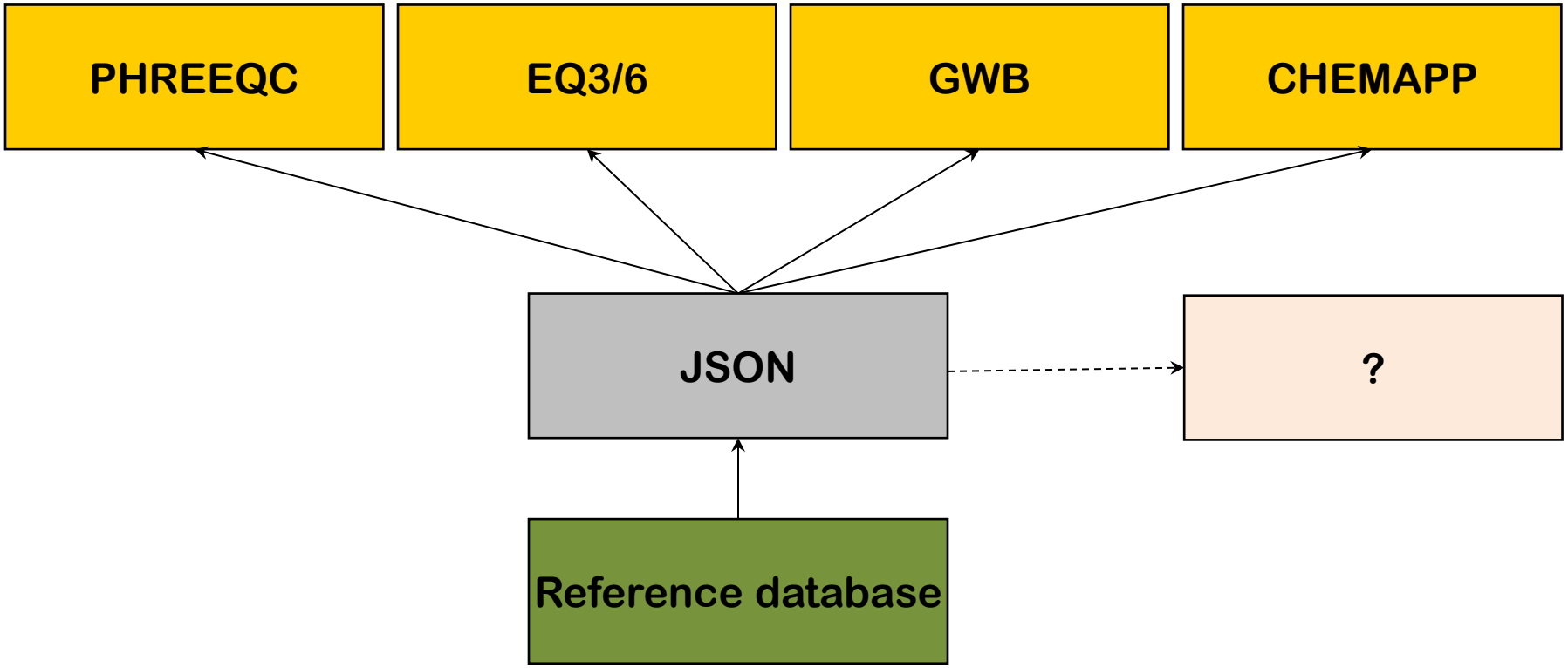
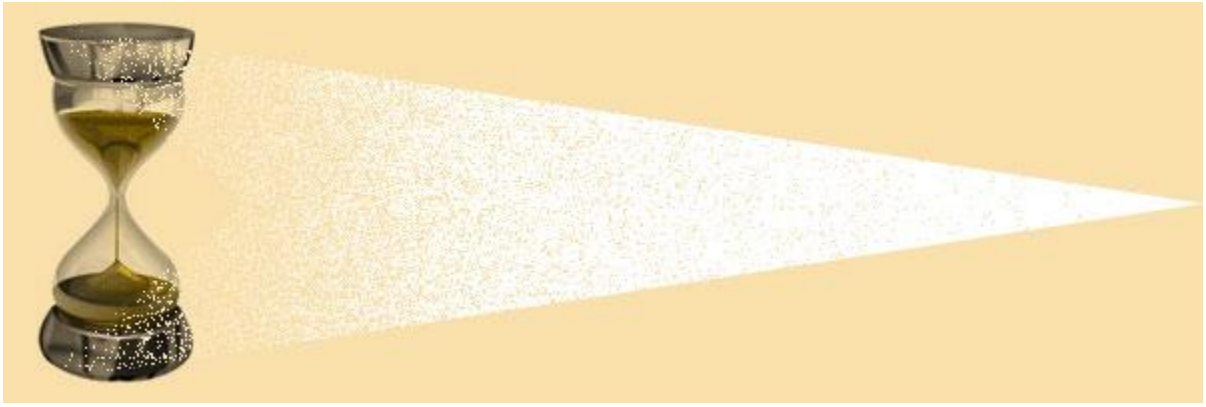
Basic idea

Thermodynamic calculations of various institutions become comparable, by creating parameter files from a common database.

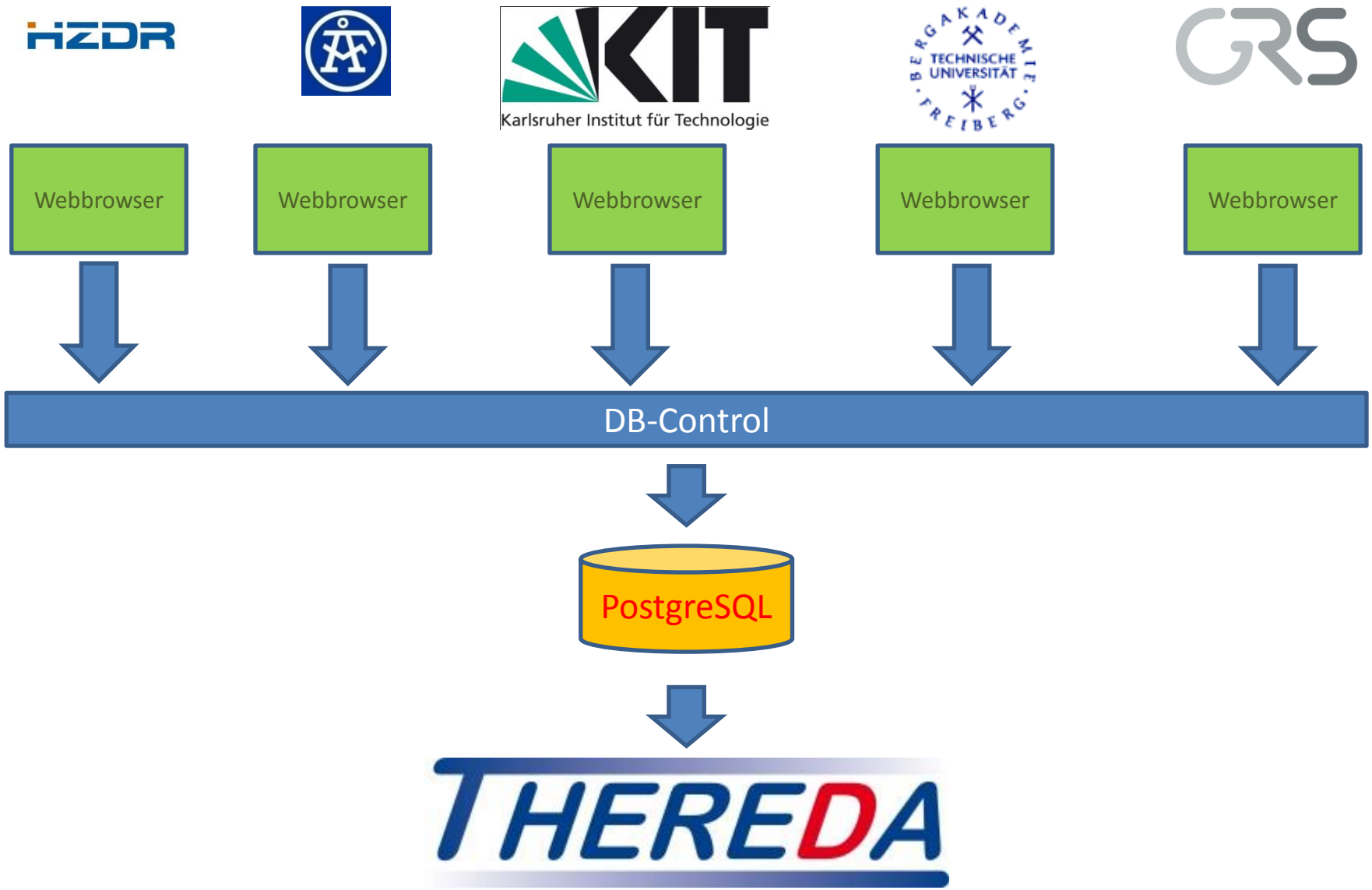


Long-term useability

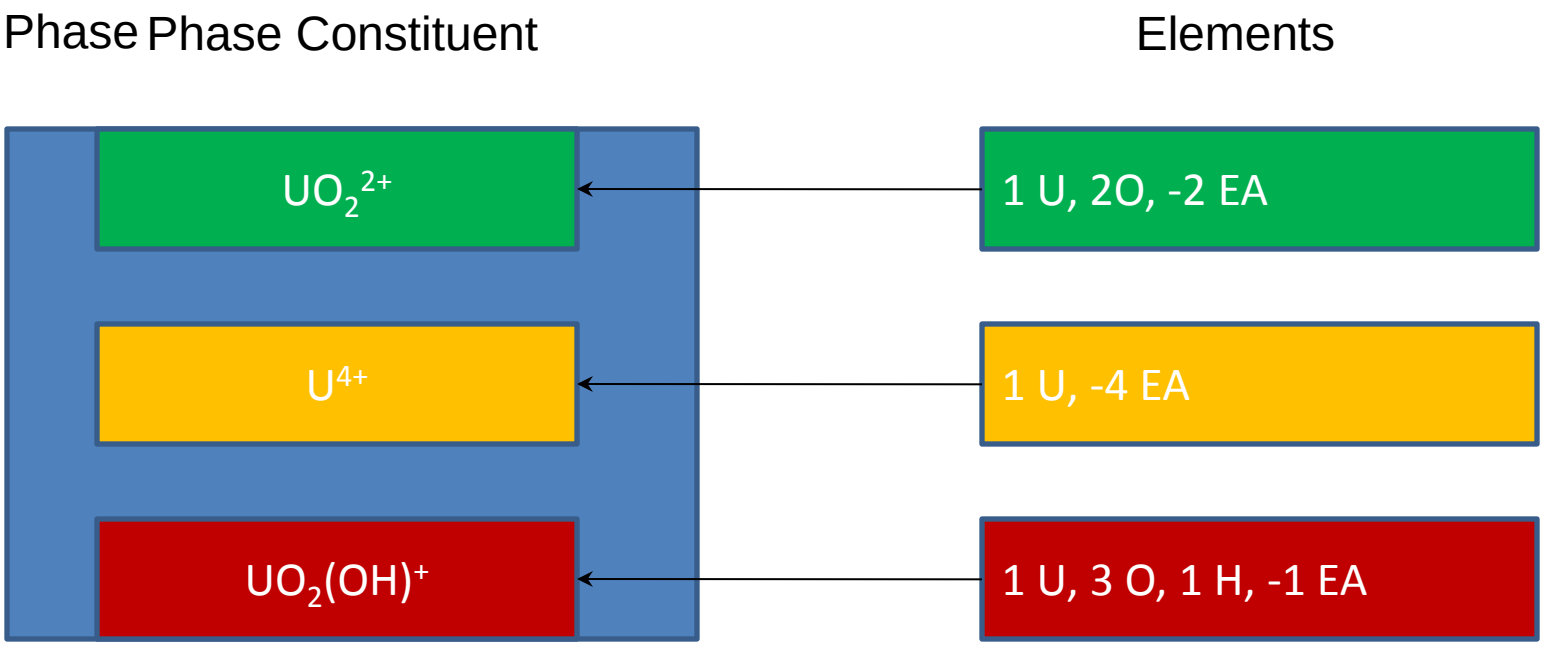
- Usage of open-source programs
  - JOOMLA!, PostgreSQL, web-browser to manipulate data
- Low degree of abstraction of the data model
- Documentation of databank structure
- Flexible databank structure
  - Various interaction models for gas- and aqueous phase and solid phases
- Backups in ASCII-format (SQL-scripts)
- Joint project of five research institutions



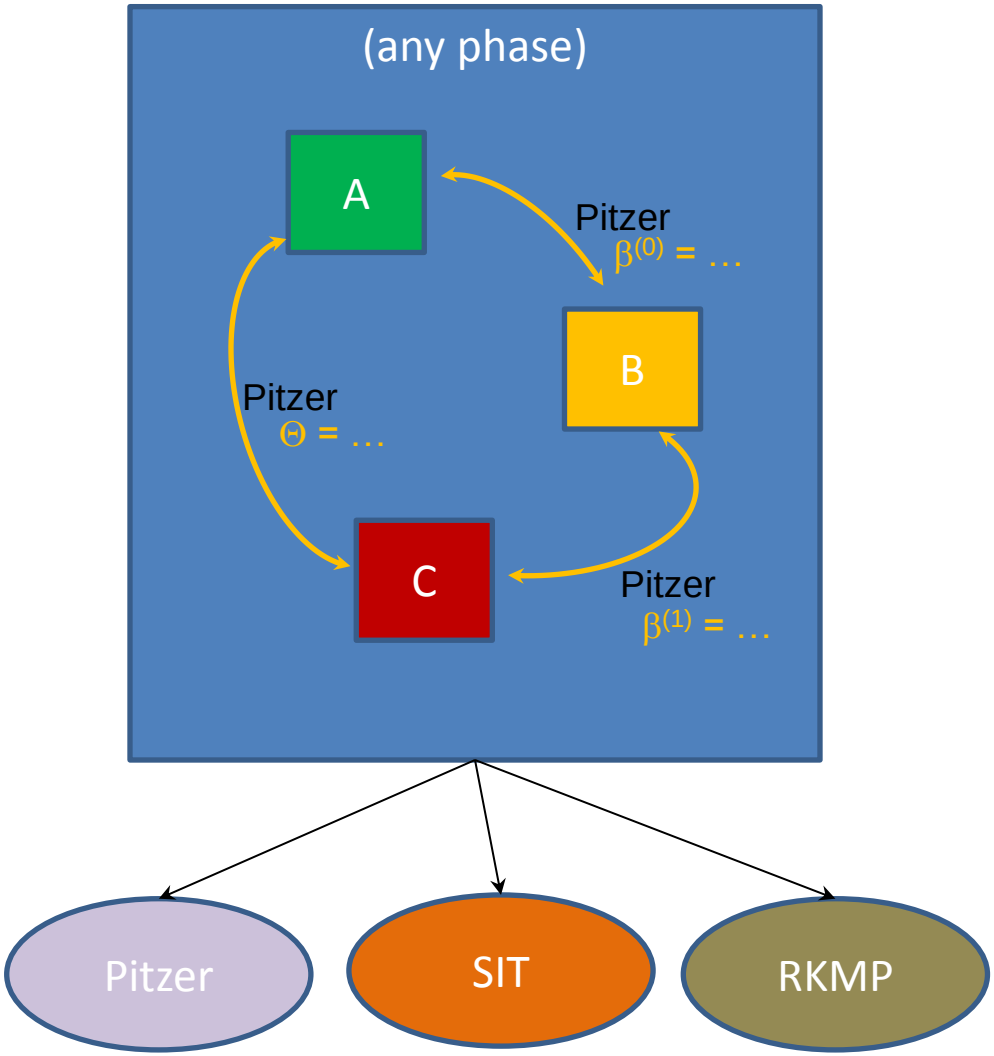
Operation of THEREDA



Data structure: Phases and Phase constituents



Data structure: non-ideal interactions



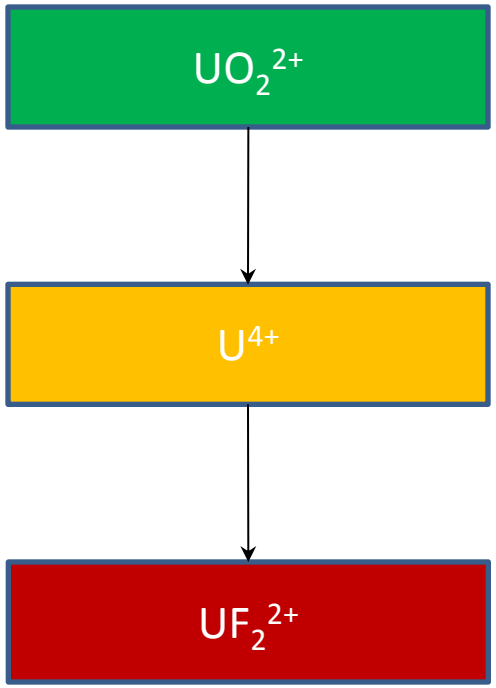
- Declaration of interaction models (Pitzer, SIT, RKMP, etc.)
- Assignment of interaction model to phase
  - Pitzer <-> aq
  - RKMP <-> (any solid mixed phase)
- Declaration of interacting phase constituents in conjunctions with a particular interaction model
- Assignment of interaction coefficients

Data structure: Phase constituent types and Reactions

Primary Master

Secondary Master

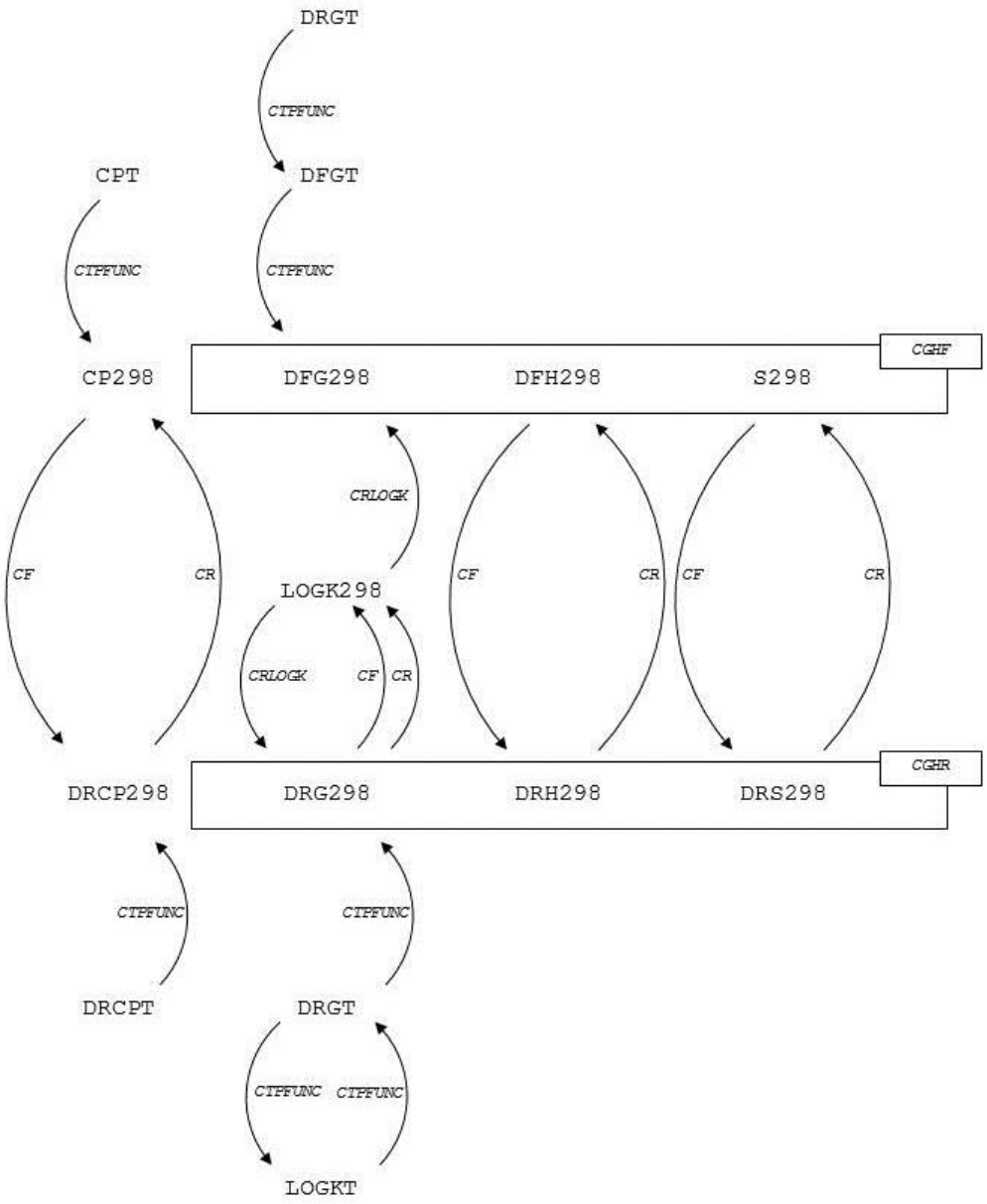
Products



Reactant	Coefficient
UF2<2+>	+1
U<4+>	-1
F<->	-2

Internal calculation scheme

- A technical framework which ensures that dependent and independent data are stored in a manner which ensures internal consistency when independent data are modified
- Editor determines, which data have to be calculated and how
- Internally, calculations are executed by SQL scripts
- Only those data are recalculated which are affected by a change



Benefit of THEREDA

- The scientist can focus on scientific issues.
- Peripheral programs assist in debugging, surveying, converting, and exporting data into ready-to-use parameter files.

Test calculations

- To be performed with parameter files created from the databank
- Demonstrate that calculational results match with lab data or expert judgment
- Make sure that results are the same with several codes (or give reasons why they cannot because of shortcomings of the code)
- Is a means of control for the user to check the accurateness of the downloaded parameter file

Test calculations released so far

- R01: **Polythermal** solubilities in selected ternary solutions with regard to the **system of oceanic salts**
- R02:
  - **Am(III), Cm(III), Nd(III)** in $MgCl_2$ and $CaCl_2$ solutions at 298.15K
  - **Am(III), Cm(III), Nd(III)** in NaCl solutions at 298.15K
- R03: Solubility in selected ternary salt solutions with regard to the system of oceanic salts in presence of **CO2 and carbonates**
- R04: Solubility and hydrolysis of **Np(V)** in NaCl solutions at 298.15K
- R05: Activities and solubilities in solutions of **Cs**, Na, K, Mg, Ca, Cl and SO_4 at 298.15 K
- R06: Solubilities and activities in solutions of metastable and stable **cement phase assemblages**
- R07: **Th(IV), Np(IV) and Pu(IV)** solubility in NaCl, $CaCl_2$, Na_2CO_3 and $KHCO_3$ solutions

These parameterfiles are ready for download to anyone interested!

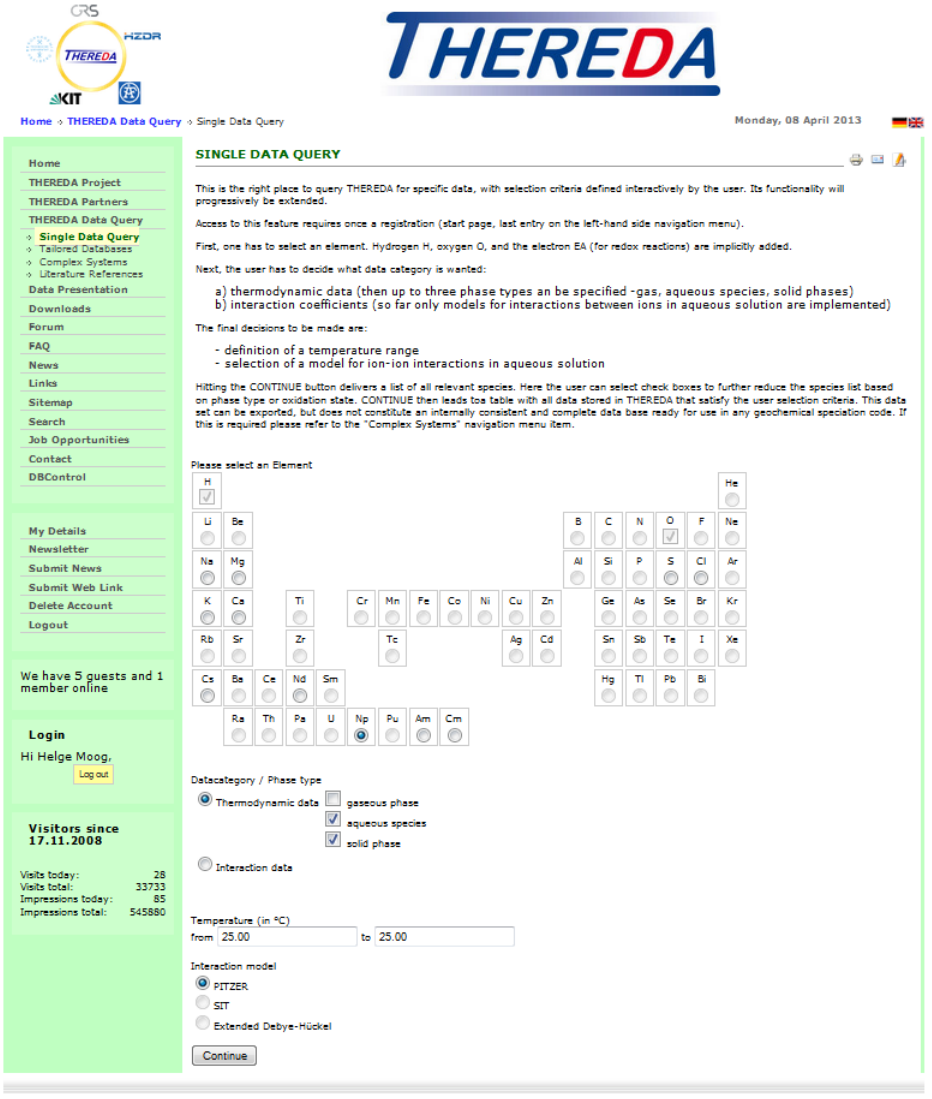
Test calculations in preparation

- (Am, Cm, Nd) – (SO_4, CO_3) – H_2O
- Na, K – PO_4 – Cl, SO_4 – H_2O
- U(+IV/+VI) solubility in Na, K, Mg, Ca – Cl, SO_4, CO_3 – Si/Al – H_2O
- Generally: solubilities of Si/Al-bearing phases at elevated temperatures
- Fe(II) – Na, K, Mg, Ca – Cl – H_2O for 25°C

Using THEREDA (1)

- Select Single Data Query
  → get equilibrium constants, Pitzer parameters
- Export as CSV or MS Excel© possible
- Select Tailored databases
  → download parameter files for
  - PHREEQC
  - EQ3/6
  - GWB
  - CHEMAPP

- **Please visit: www.thereda.de**



Using THEREDA (2): JSON

- Export data in a gene
- “JavaScript Object N
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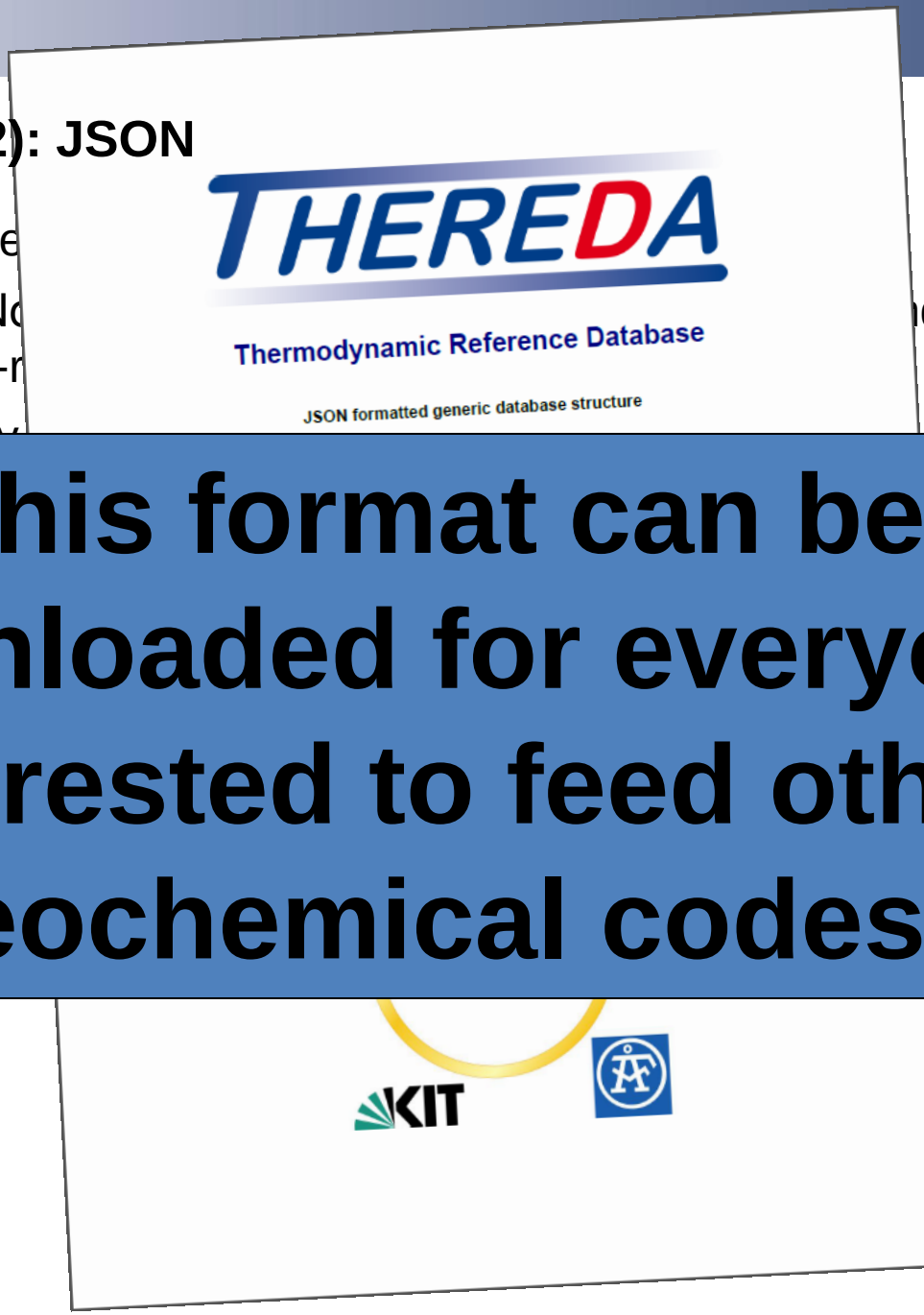
- P



This format can be downloaded for everyone interested to feed other geochemical codes!

Current activities / Outlook (1)

- THEREDA has been extended to hold **surface complexation data**. Further activities will include the implementation in DB-Control.
- Implementation of a model for **non-ideal gases**
- Extension of some data-sets to **higher temperatures**
- Possibly addition of **another supported code**

Current activities / Outlook (2) – Surface Complexation Data

- Intensive literature search & critical assessment of Surface Complexation data → {RES3T} → Generation of “Recommended Values” → THEREDA
- Consistent with Extended-Debye-Hückel data
- Mineral specific information:
  - Specific Surface Area [m^2/g]
  - Surface Site Density [nm^{-2}]
  - Capacitance values [F/m^2]
- Mineral-Ligand-combinations data - Surface Protolysis (p*K*) & Surface Complex Formation constants (log*K*) for:
  - Diffuse Double-Layer Model
  - Constant Capacitance Model
  - Non-electrostatic Model
  - 1pK-Basic Stern Model
- Test calculations including comparisons with measured values from literature
- Data releases with “ready-to-use” parameter files for SCM-supporting geochemical codes
- Currently Surface complexation Modeling data will be collected for 298.15 K only, but adaptation to temperature-dependent functions is possible.

Conclusion

- THEREDA offers a web-based, thermodynamic reference database **for high-saline aqueous systems** in equilibrium with **nuclear waste** forms in Germany
- **Joint-project** of five research institutions
- Access to **code-specific parameter files**

Acknowledgements

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Thank you very much for your attention!