A Contribution to the Long-Term Safety of Repositories for Nuclear and Non-Nuclear Waste

Helge C. Moog

What you should know after this presentation

Thermodynamic equilibrium modeling – what do we need it for?

What is necessary to perform a calculation

Our concern: reliable model calculations as contribution to safety analysis

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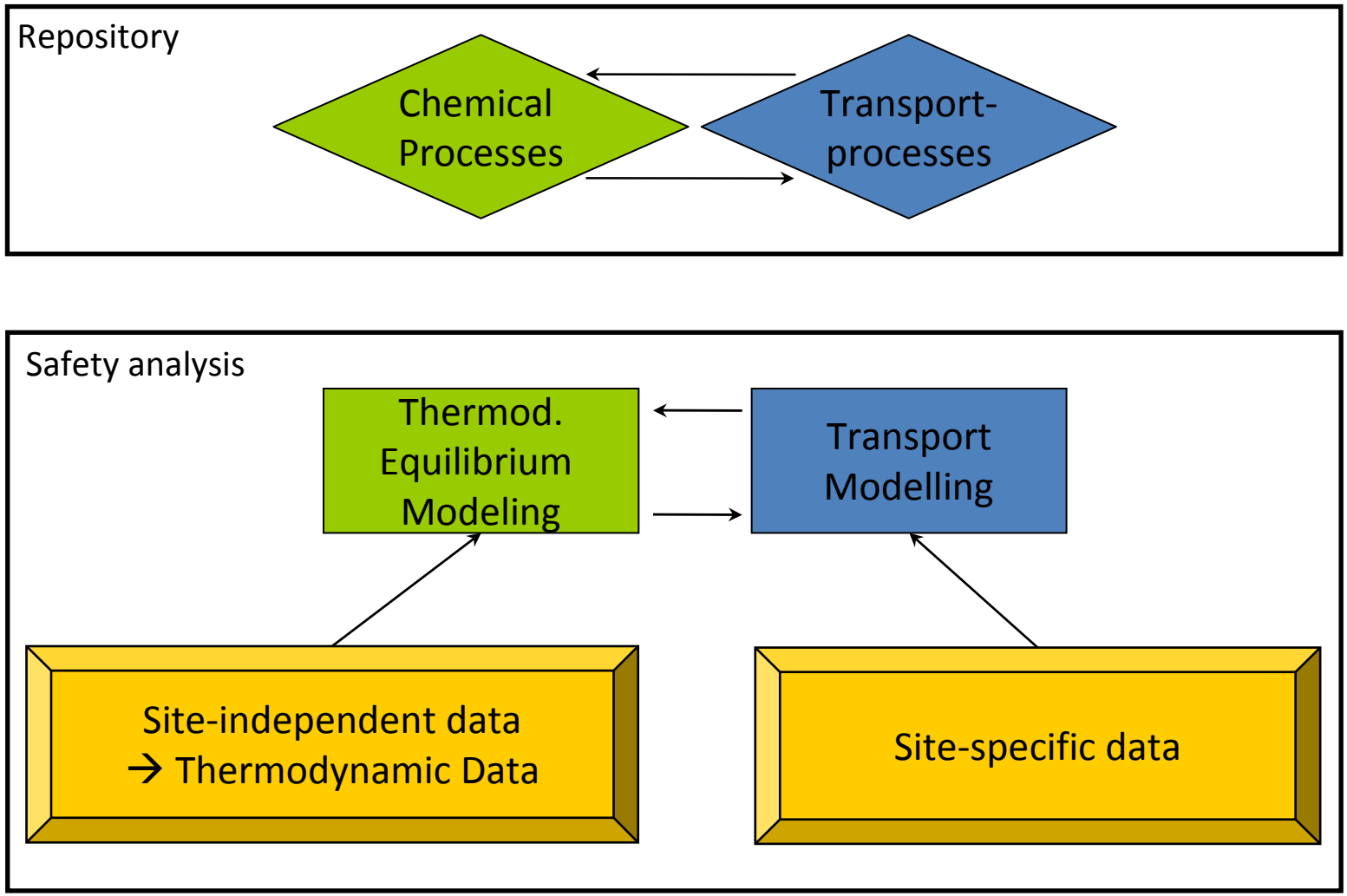
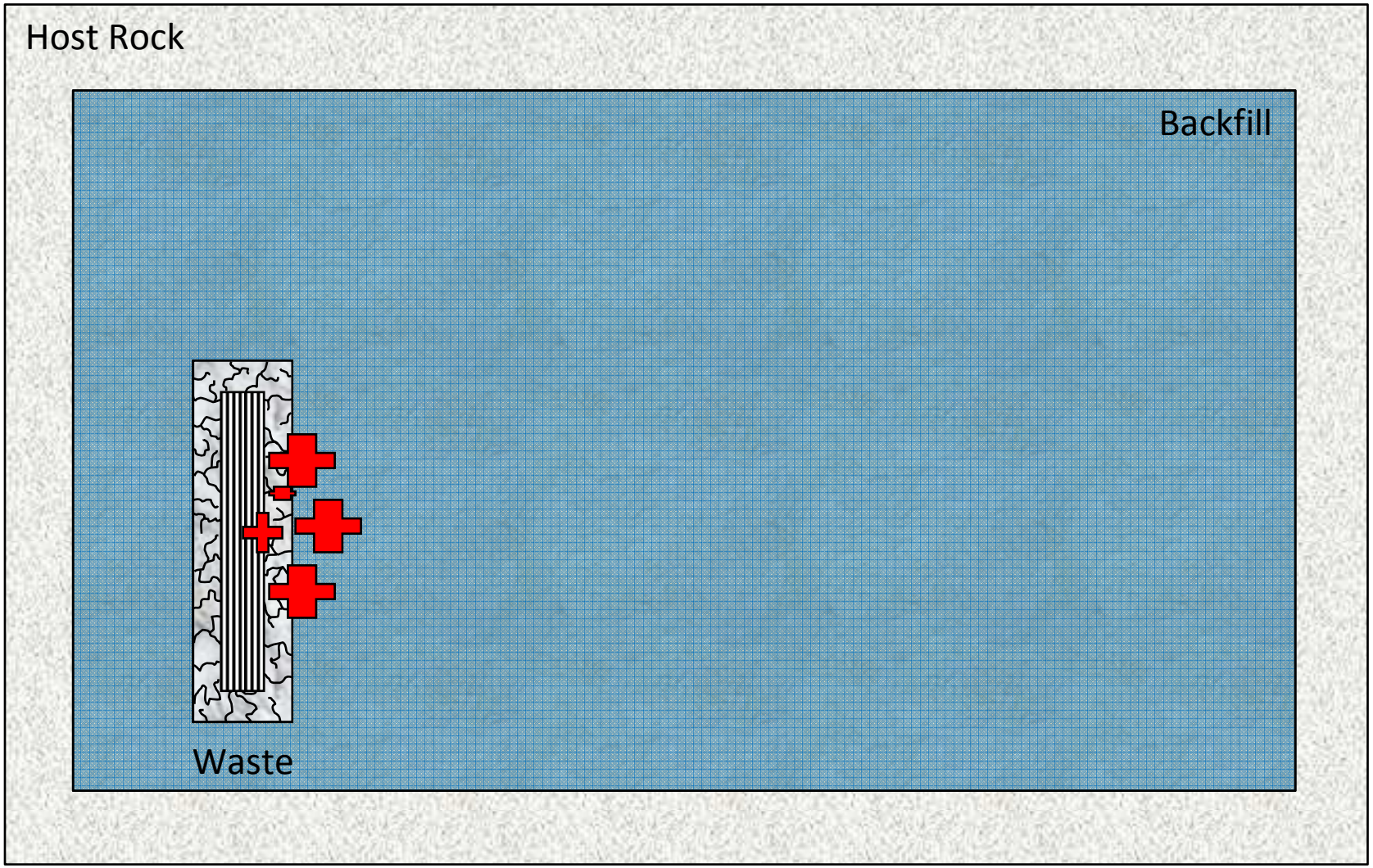
- THEREDA – a databank for thermodynamic reference data
  - Technology
  - QS-Elements
  - Future prospects

Thermodynamic equilibrium modeling – what do we need it for?



What are „chemical prozesses“?



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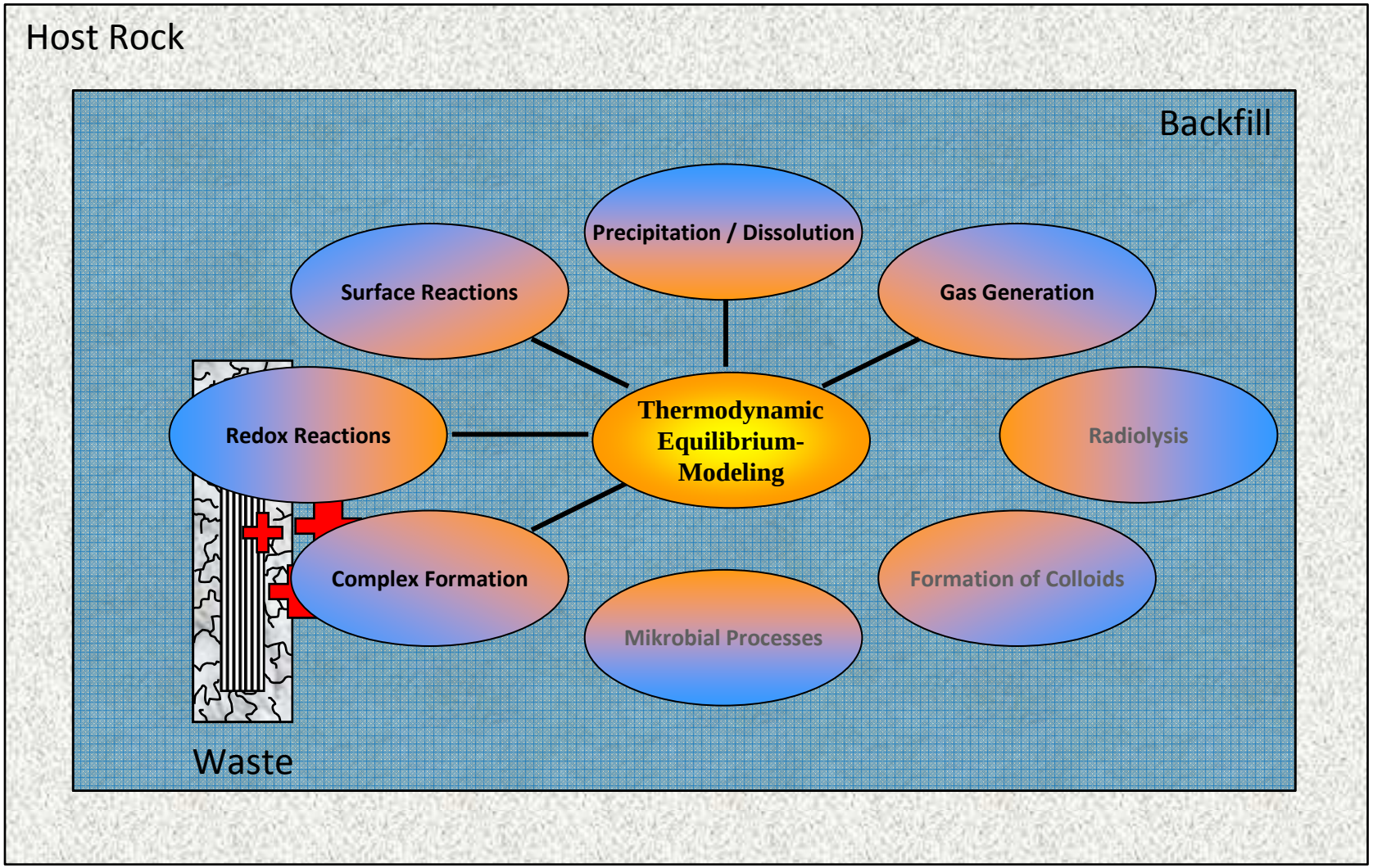
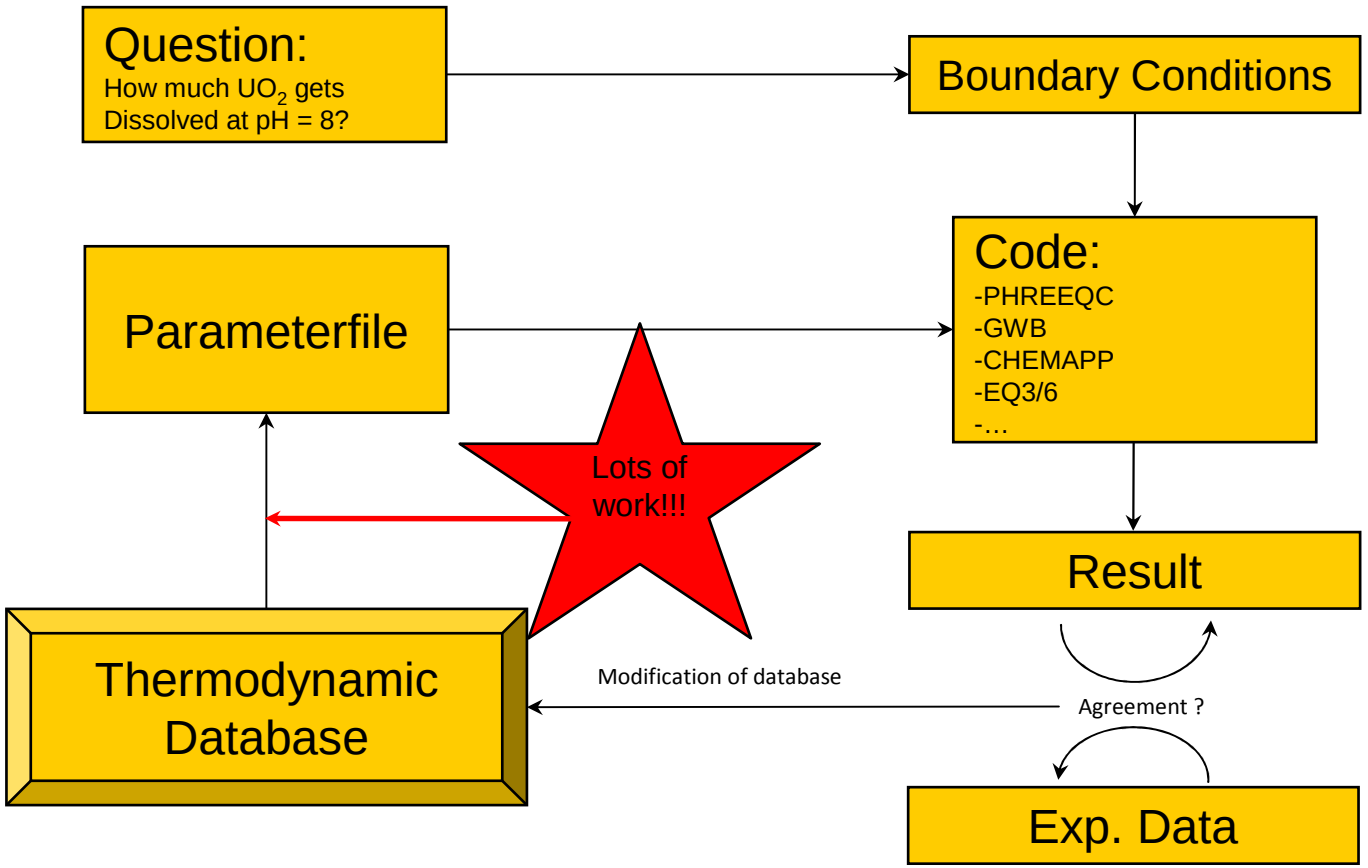


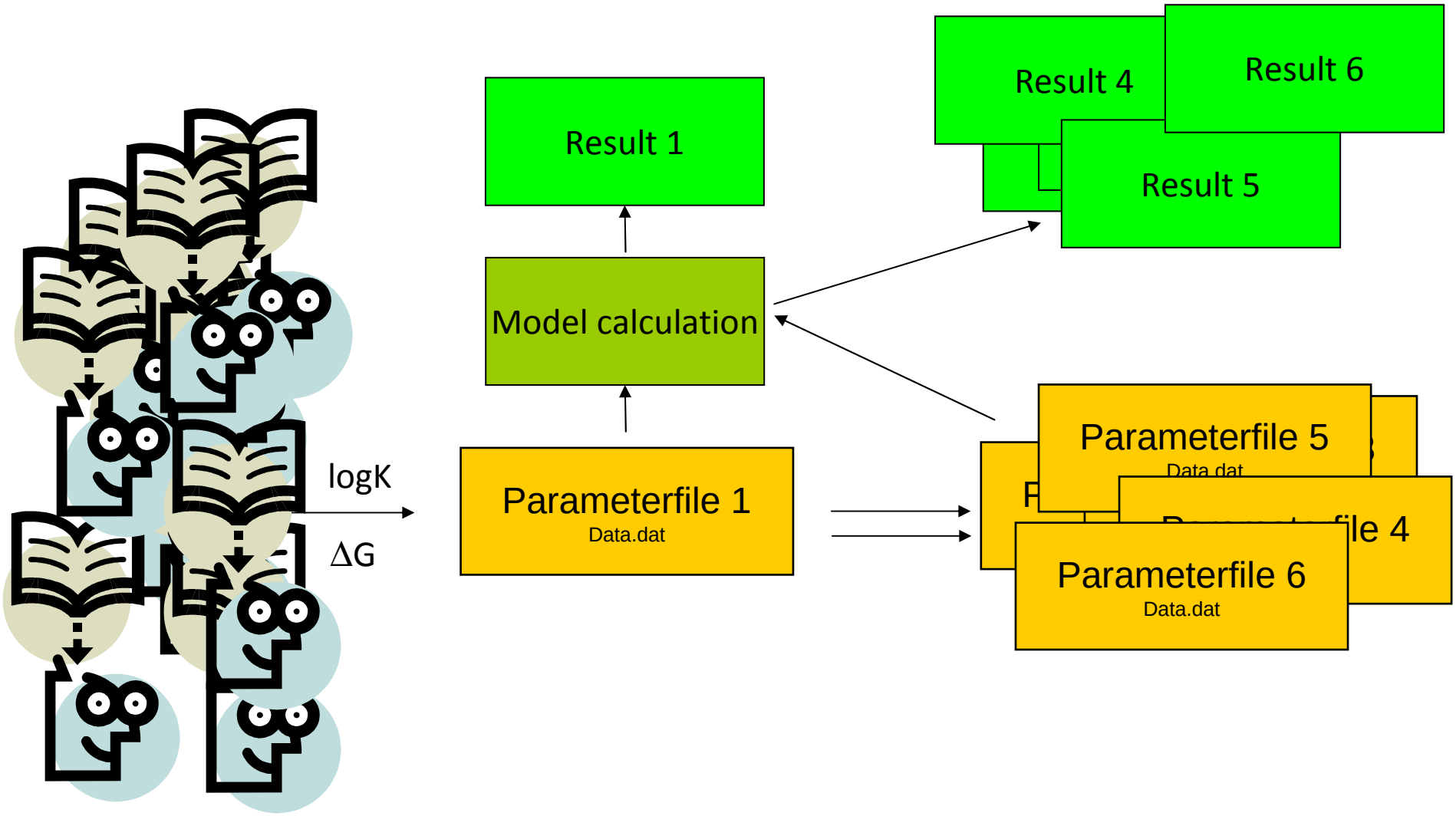
What do we need to perform a thermodynamic equilibrium calculation?



Our concern:
<u>Reliable</u> calculations as contribution to safety analysis

Which „parameterfile“ are we talking about, when it comes safety analysis??!

Thermodynamic Equilibrium Modeling in practise

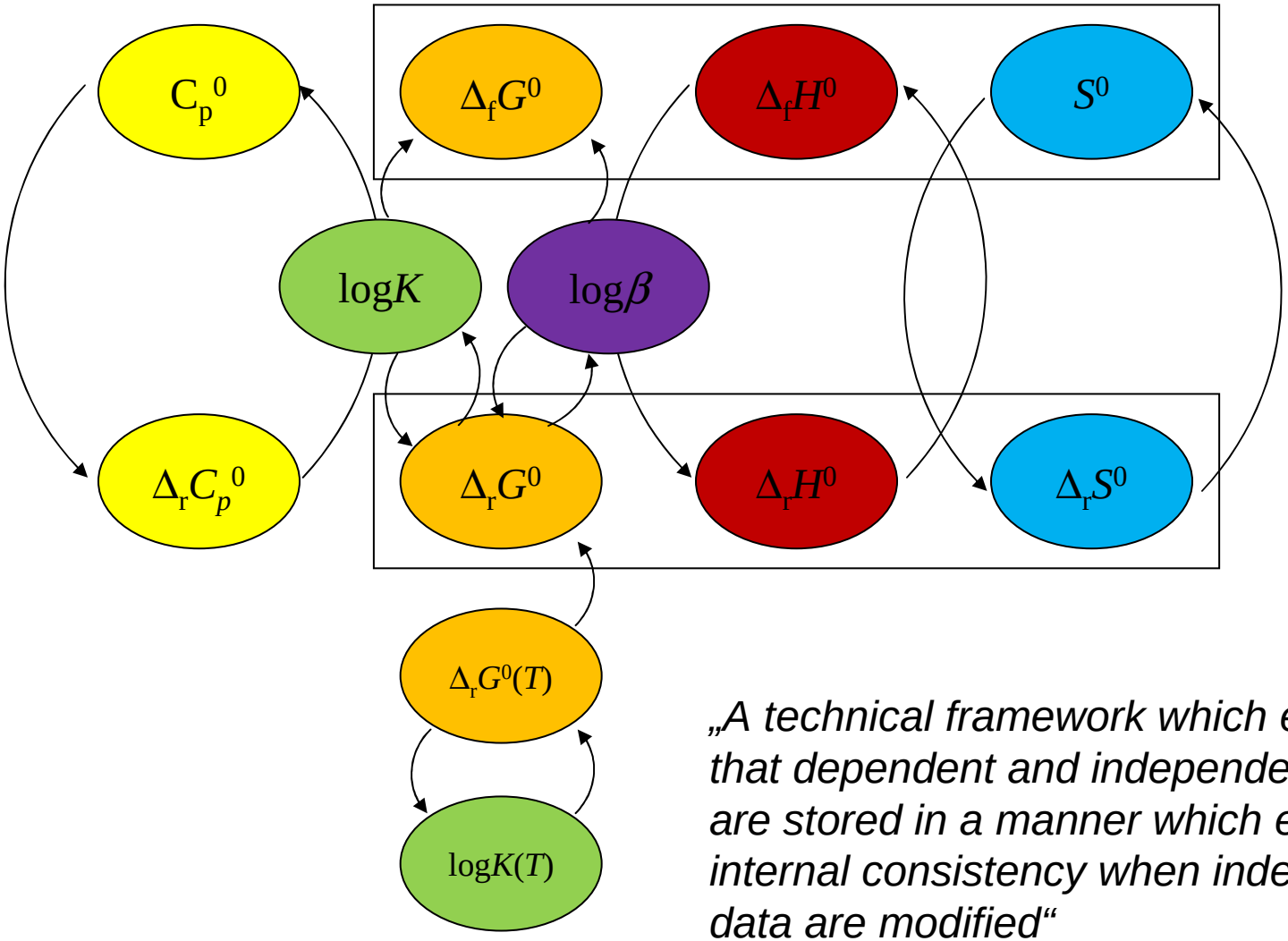


Thermodynamic Equilibrium Modeling in practise

Which result is correct????!

What do we mean by „thermodynamic database“?

Consistency of data



„A technical framework which ensures that dependent and independent data are stored in a manner which ensures internal consistency when independent data are modified“

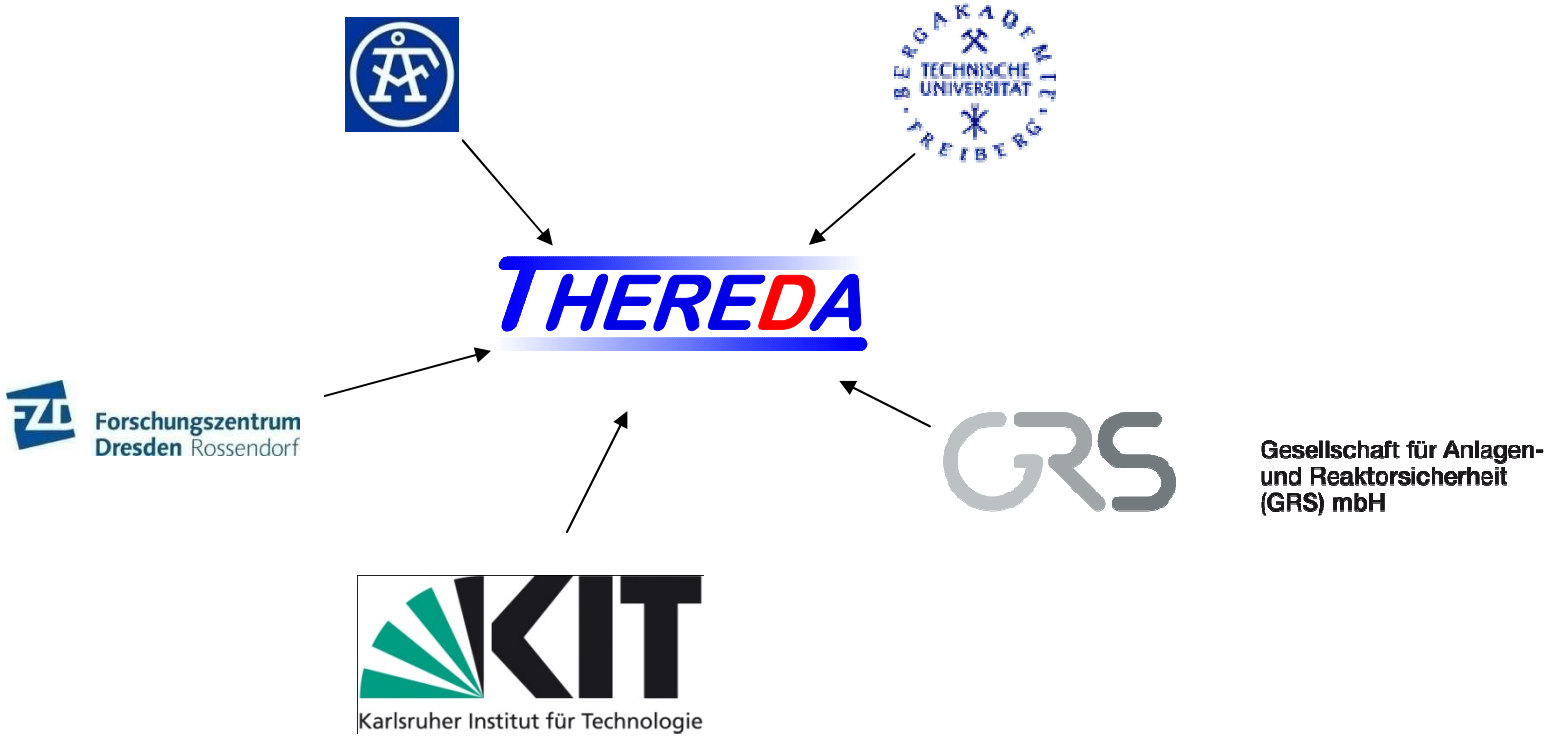
Thermodynamic equilibrium modeling for nuclear and non-nuclear modeling in the future

Creation of a consistent, mutually agreed and quality-assured reference database
Joining competences for a common sake
Easy access via the world wide web
Making available ready-to-use parameter files

h1 Modellierung des Gesamtprozesses nur so gut wie die Modellierungsqualität der Einzelprozesse

has; 14.04.2005

Thermodynamic equilibrium modeling for nuclear and non-nuclear modeling in the future



h2 Modellierung des Gesamtprozesses nur so gut wie die Modellierungsqualität der Einzelprozesse

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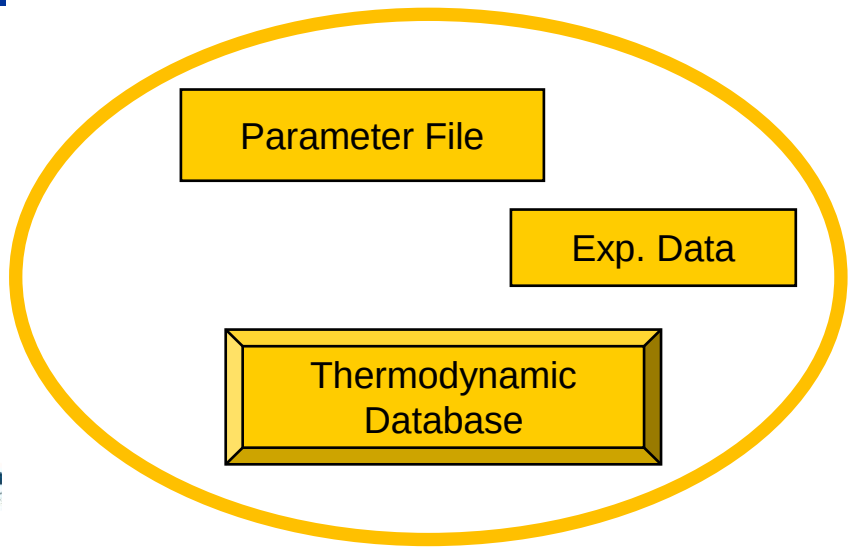
Thermodynamic equilibrium modeling for nuclear and non-nuclear modeling in the future



Forschungszentrum Dresden Rossendorf

Karlsruher Institut für Technologie

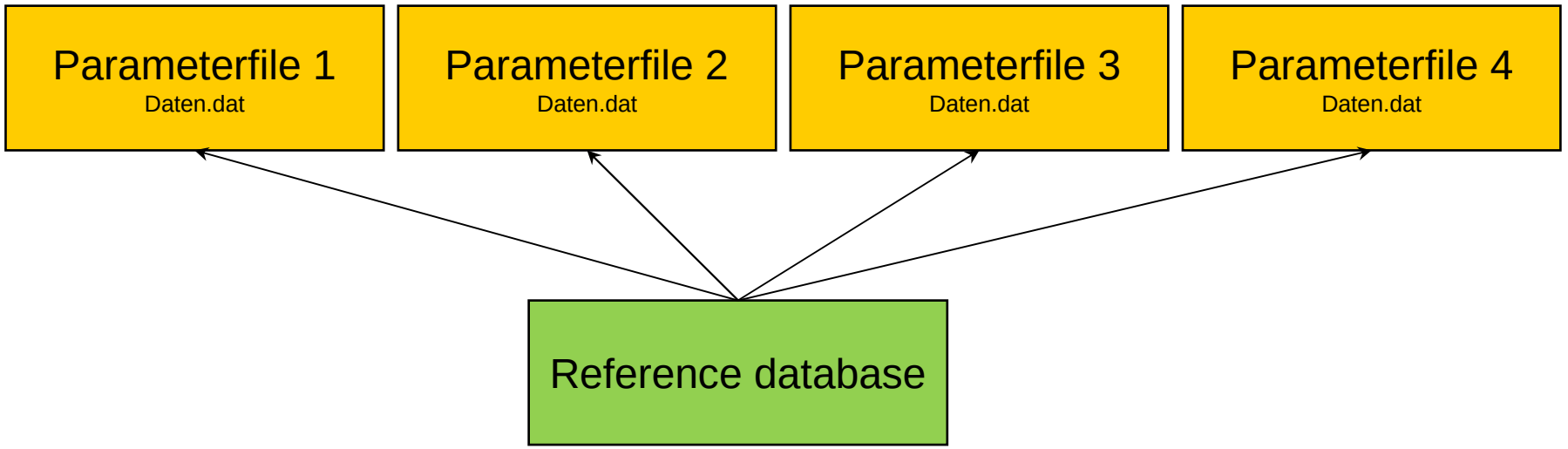
Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) mbH

h3 Modellierung des Gesamtprozesses nur so gut wie die Modellierungsqualität der Einzelprozesse

has; 14.04.2005

Basic idea

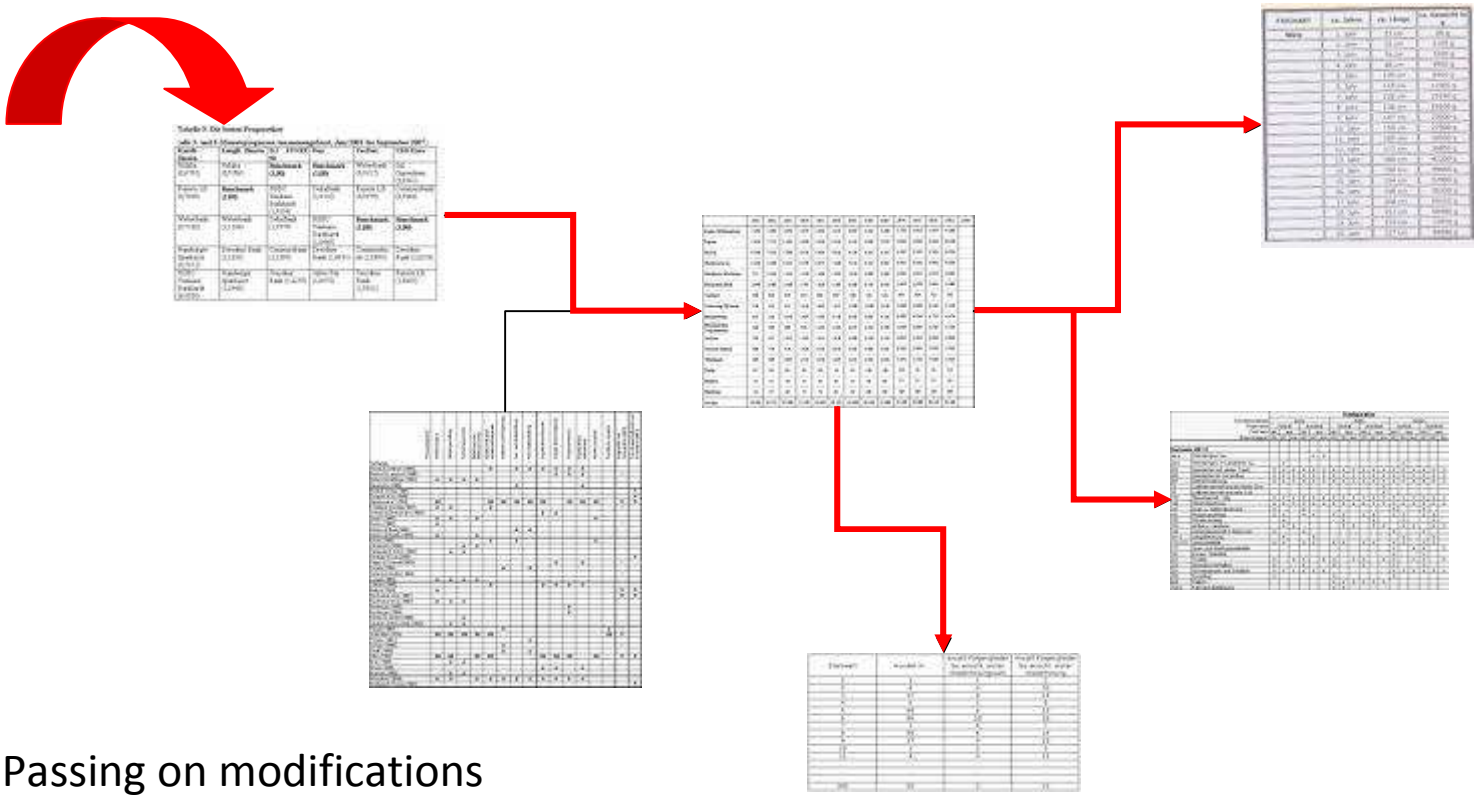
Thermodynamic calculations of various institutions become compareable, by creating parameter files from a common database.



Further project data

- Phase I: 2006-07 bis 2010-03
- Phase II: 2009-10 bis 2013-03
- Funded by two Ministries: BMWi und BMBF, Funding approved by BfS (Federal Office for Radiation Protection) at 3.5. (Letter of Intent)

Technical Basics



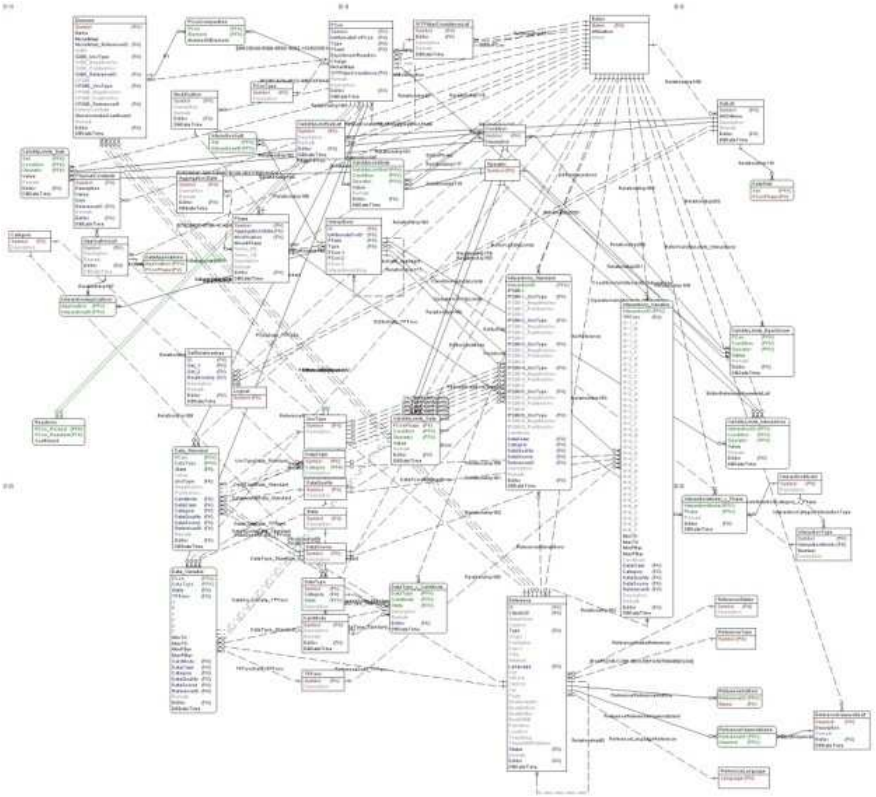
Passing on modifications

Organised in Tables

Referential Integrity

Technical Basics

- Datenbanksystem = PostgreSQL
  - Relations: >56
  - Attributes: >370
  - Constraints: >120



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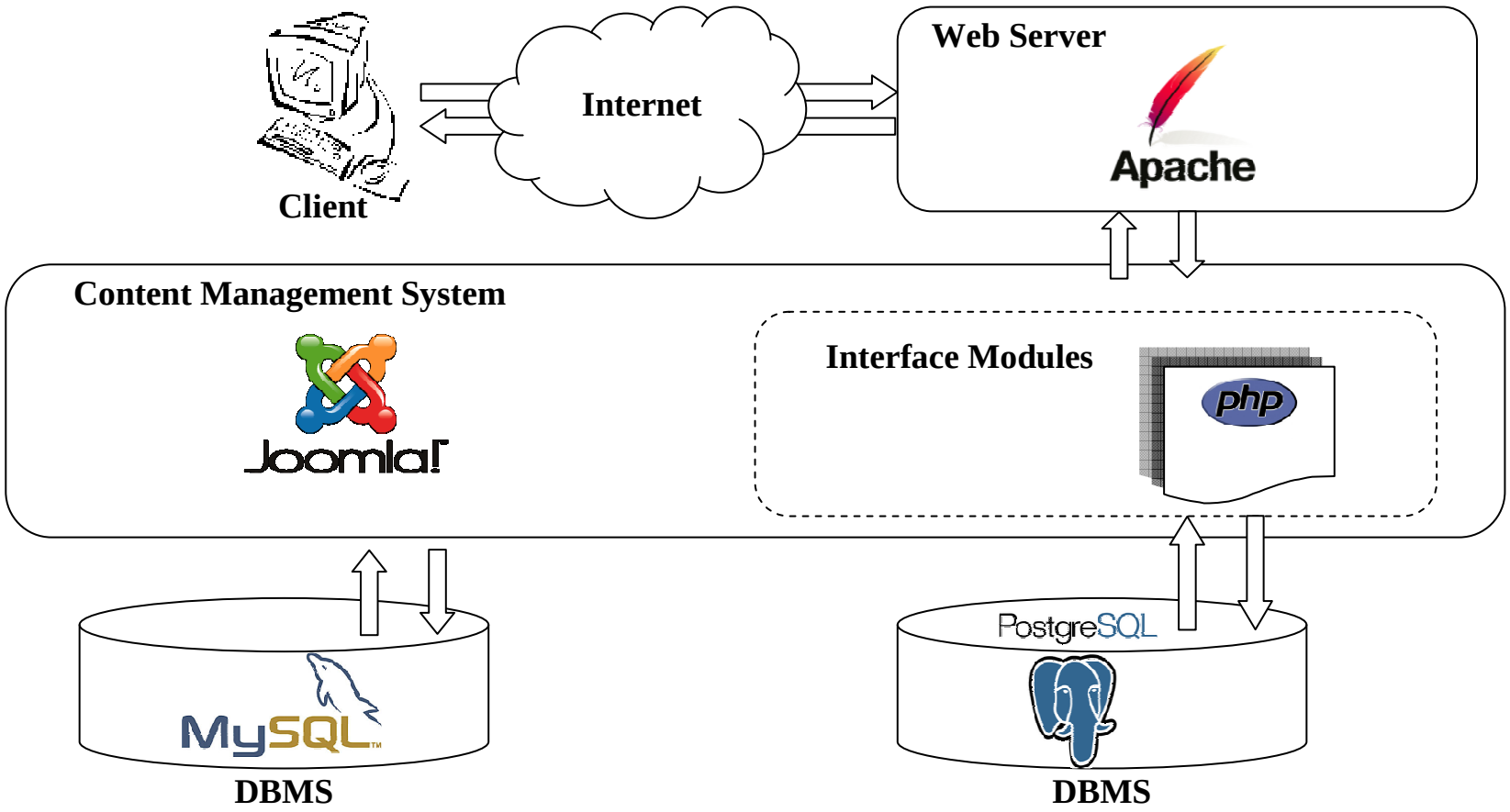
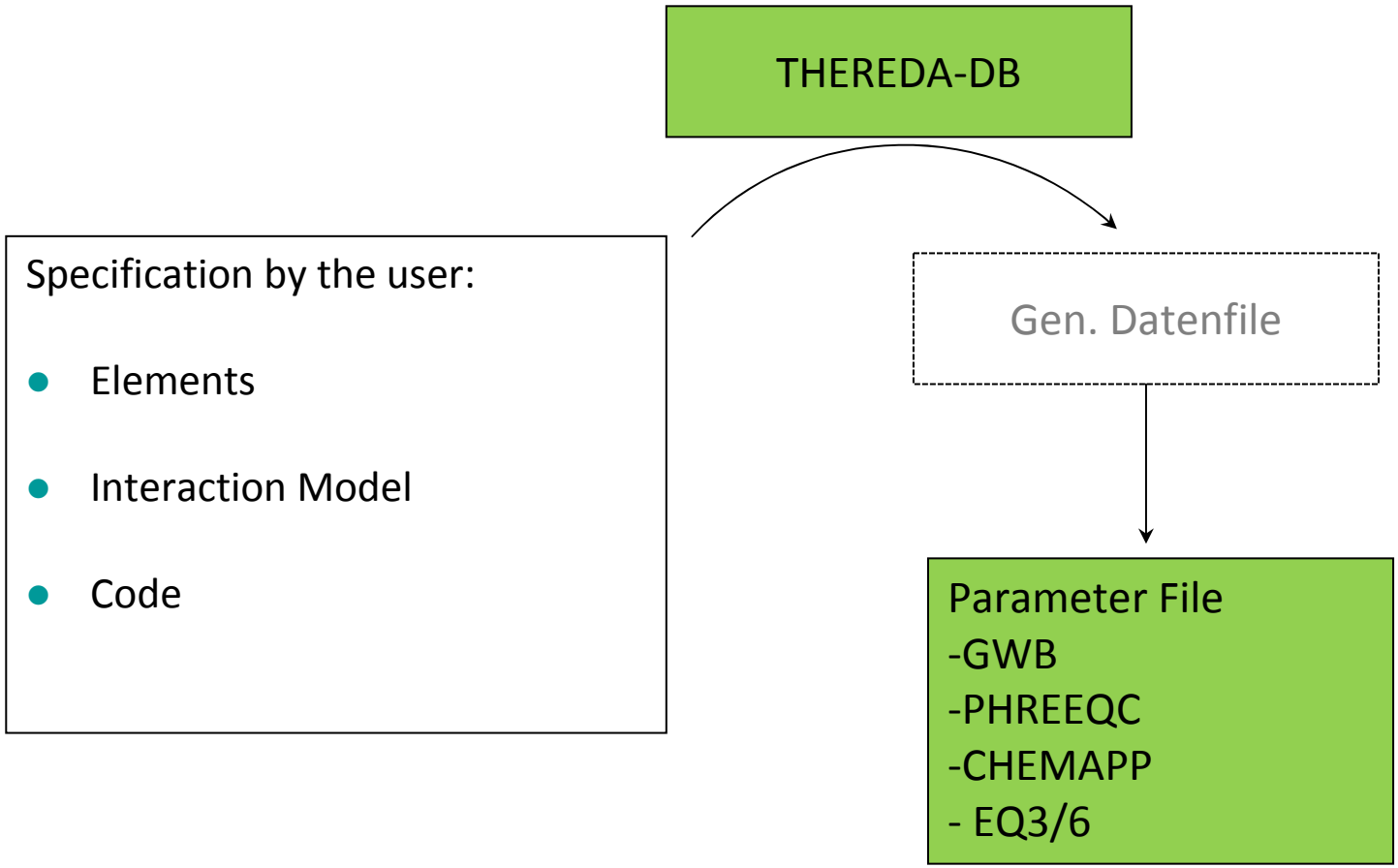


How it is used

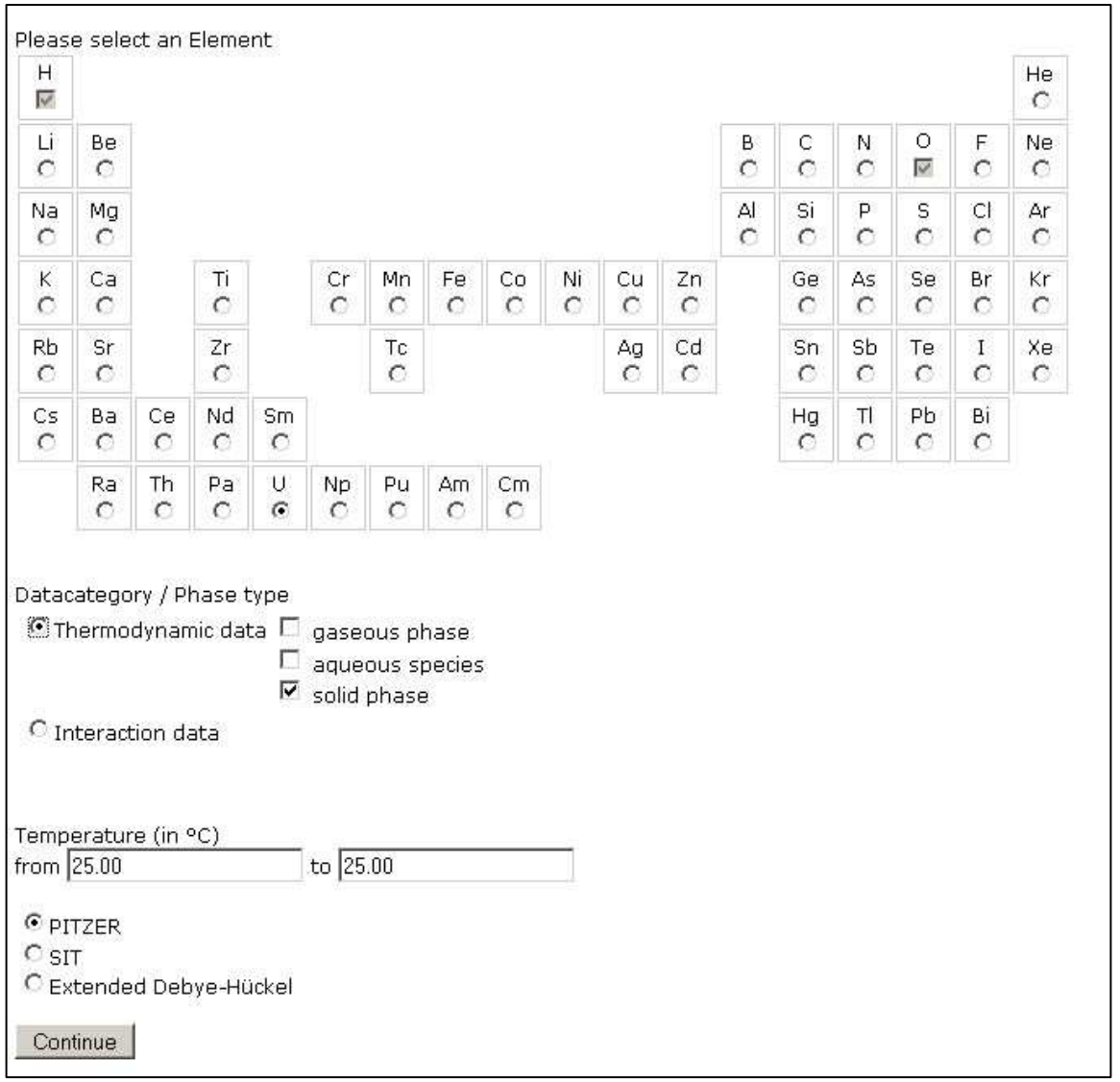


Access via World Wide Web

www.thereda.de

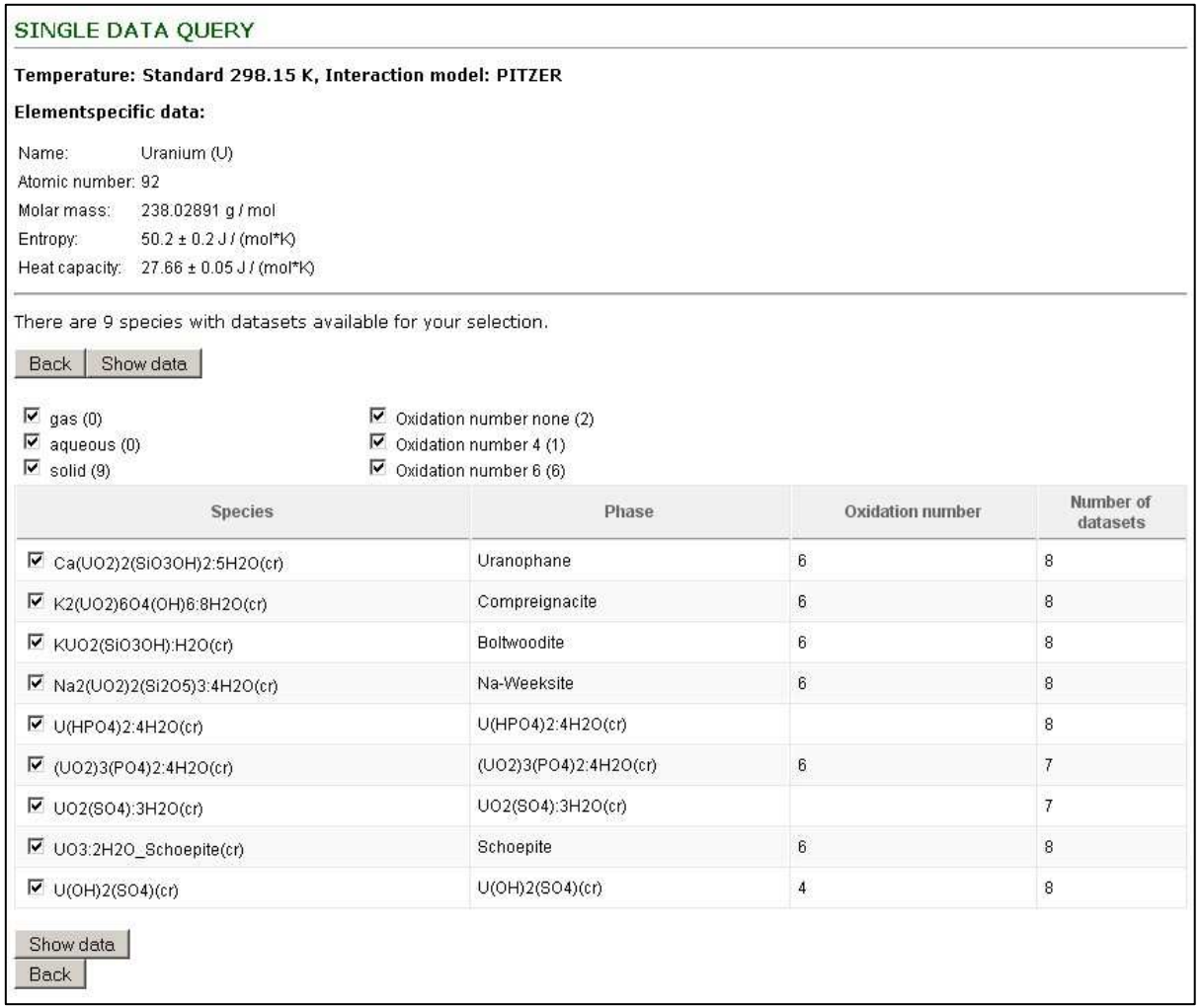
Access via World Wide Web

- Query is performed interactively with the user
- Thermodynamic data and interaction coefficients



Access via World Wide Web

- Phase constituents grouped by phases and oxidation number of central element
- Thermodynamic data of pure elements
- With [Show Data] display of detailed data



SINGLE DATA QUERY

Temperature: Standard 298.15 K, Interaction model: PITZER

Elementspecific data:

Name:	Uranium (U)
Atomic number:	92
Molar mass:	238.02891 g / mol
Entropy:	50.2 ± 0.2 J / (mol*K)
Heat capacity:	27.66 ± 0.05 J / (mol*K)

There are 9 species with datasets available for your selection.

Back | Show data

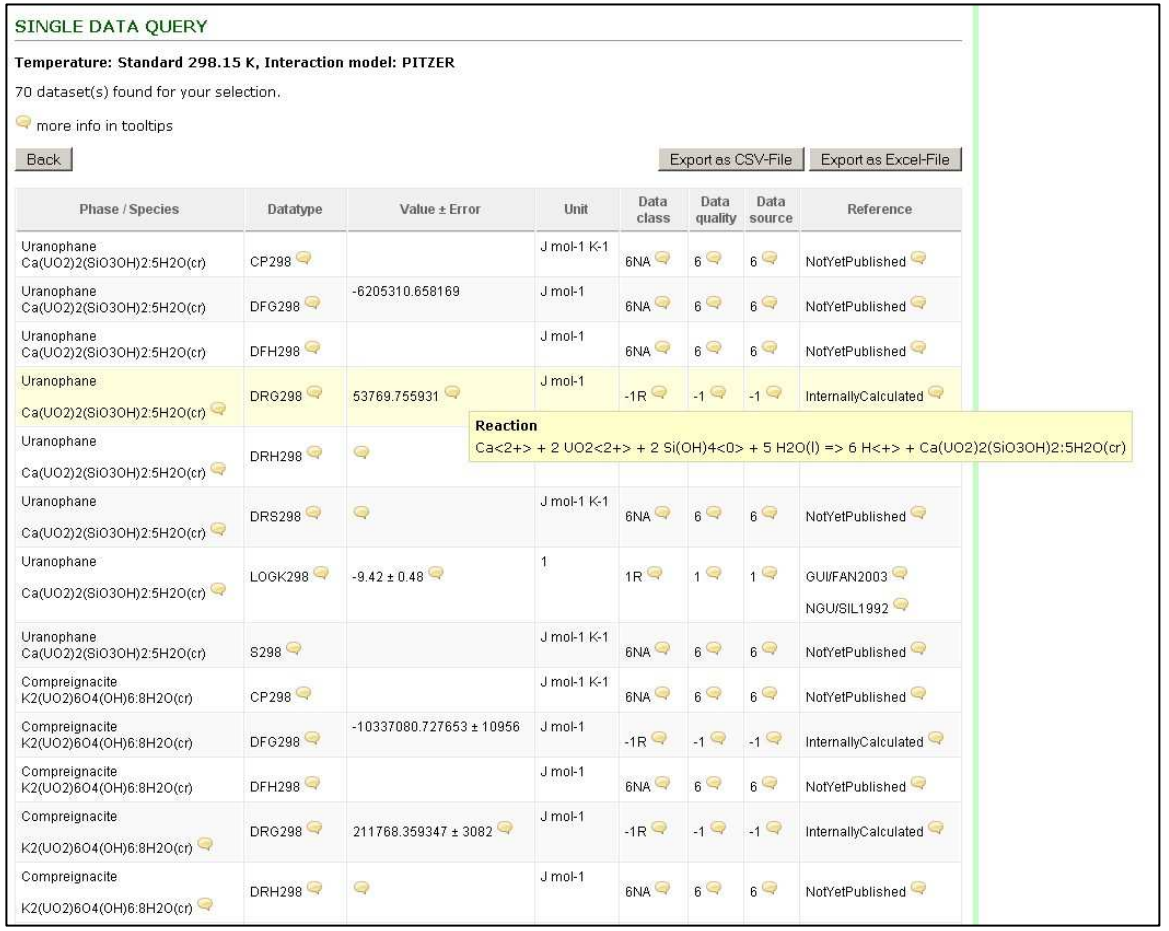
☑ gas (0)
☑ aqueous (0)
☑ solid (9)
☑ Oxidation number none (2)
☑ Oxidation number 4 (1)
☑ Oxidation number 6 (6)

Species	Phase	Oxidation number	Number of datasets
☑ Ca(UO2)2(SiO3OH)2:5H2O(cr)	Uranophane	6	8
☑ K2(UO2)6O4(OH)6:8H2O(cr)	Compreignacite	6	8
☑ KUO2(SiO3OH):H2O(cr)	Boltwoodite	6	8
☑ Na2(UO2)2(Si2O5)3:4H2O(cr)	Na-Weeksite	6	8
☑ U(HPO4)2:4H2O(cr)	U(HPO4)2:4H2O(cr)		8
☑ (UO2)3(PO4)2:4H2O(cr)	(UO2)3(PO4)2:4H2O(cr)	6	7
☑ UO2(SO4):3H2O(cr)	UO2(SO4):3H2O(cr)		7
☑ UO3:2H2O_Schoepite(cr)	Schoepite	6	8
☑ U(OH)2(SO4)(cr)	U(OH)2(SO4)(cr)	4	8

Show data
Back

Access via World Wide Web

- Export of data as CSV or MS-Excel©
- Tool-Tips with additional information (formation reaction, classification of data, references)



SINGLE DATA QUERY

Temperature: Standard 298.15 K, Interaction model: PITZER

70 dataset(s) found for your selection.

more info in tooltips

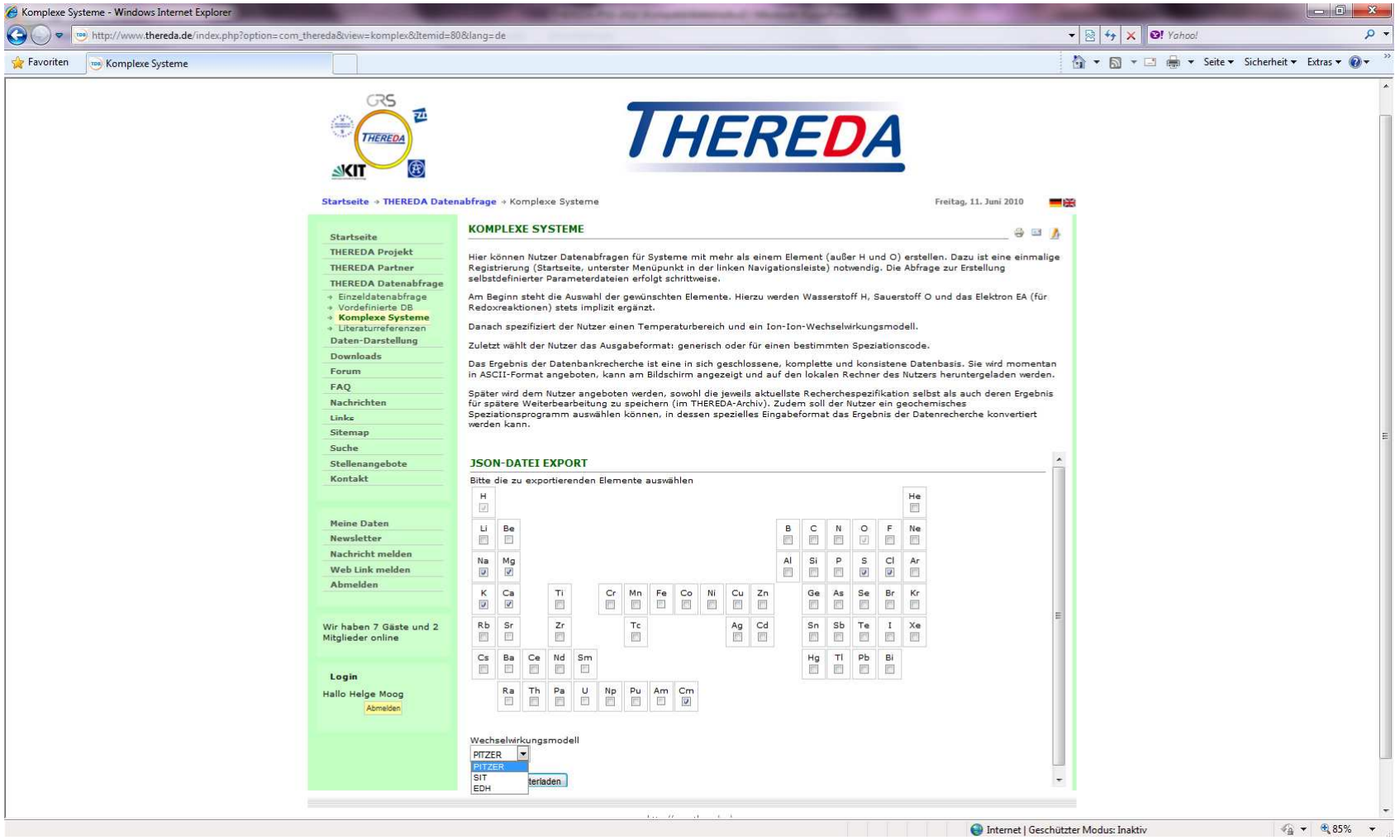
Back | Export as CSV-File | Export as Excel-File

Phase / Species	Datatype	Value ± Error	Unit	Data class	Data quality	Data source	Reference
Uranophane Ca(UO2)2(SiO3OH)2:5H2O(cr)	CP298		J mol-1 K-1	6NA	6	6	NotYetPublished
Uranophane Ca(UO2)2(SiO3OH)2:5H2O(cr)	DFG298	-6205310.658169	J mol-1	6NA	6	6	NotYetPublished
Uranophane Ca(UO2)2(SiO3OH)2:5H2O(cr)	DFH298		J mol-1	6NA	6	6	NotYetPublished
Uranophane Ca(UO2)2(SiO3OH)2:5H2O(cr)	DRG298	53769.755931	J mol-1	-1R	-1	-1	InternallyCalculated
Uranophane Ca(UO2)2(SiO3OH)2:5H2O(cr)	DRH298						
Uranophane Ca(UO2)2(SiO3OH)2:5H2O(cr)	DRS298		J mol-1 K-1	6NA	6	6	NotYetPublished
Uranophane Ca(UO2)2(SiO3OH)2:5H2O(cr)	LOGK298	-9.42 ± 0.48	1	1R	1	1	GUI/FAN2003 NGU/SIL1992
Uranophane Ca(UO2)2(SiO3OH)2:5H2O(cr)	S298		J mol-1 K-1	6NA	6	6	NotYetPublished
Compreignacite K2(UO2)6O4(OH)6:8H2O(cr)	CP298		J mol-1 K-1	6NA	6	6	NotYetPublished
Compreignacite K2(UO2)6O4(OH)6:8H2O(cr)	DFG298	-10337080.727653 ± 10956	J mol-1	-1R	-1	-1	InternallyCalculated
Compreignacite K2(UO2)6O4(OH)6:8H2O(cr)	DFH298		J mol-1	6NA	6	6	NotYetPublished
Compreignacite K2(UO2)6O4(OH)6:8H2O(cr)	DRG298	211768.359347 ± 3082	J mol-1	-1R	-1	-1	InternallyCalculated
Compreignacite K2(UO2)6O4(OH)6:8H2O(cr)	DRH298		J mol-1	6NA	6	6	NotYetPublished

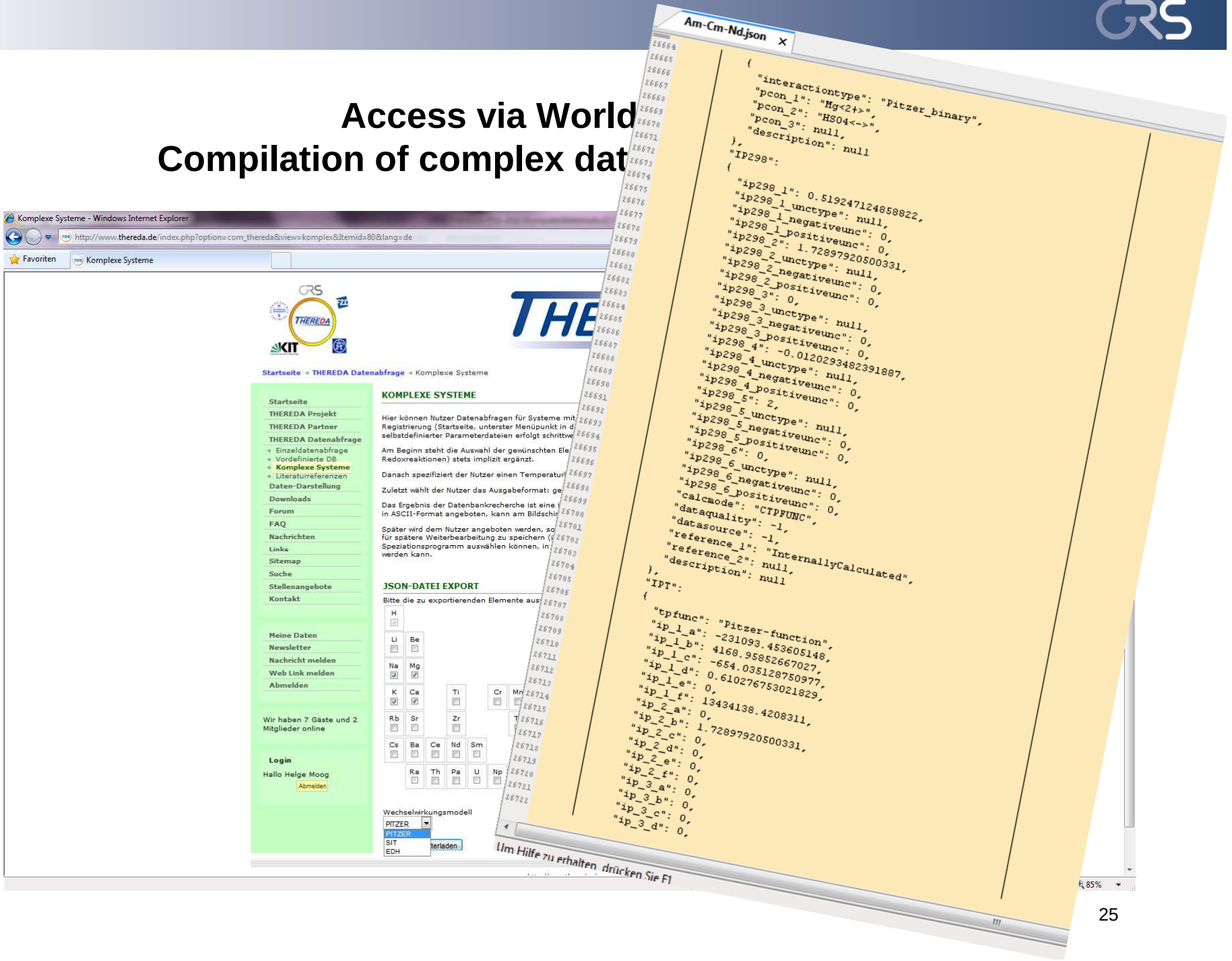
Reaction

Ca<2+> + 2 UO2<2+> + 2 Si(OH)4<0> + 5 H2O(l) => 6 H<+> + Ca(UO2)2(SiO3OH)2:5H2O(cr)

Access via World Wide Web
Compilation of complex datasets (generic format)



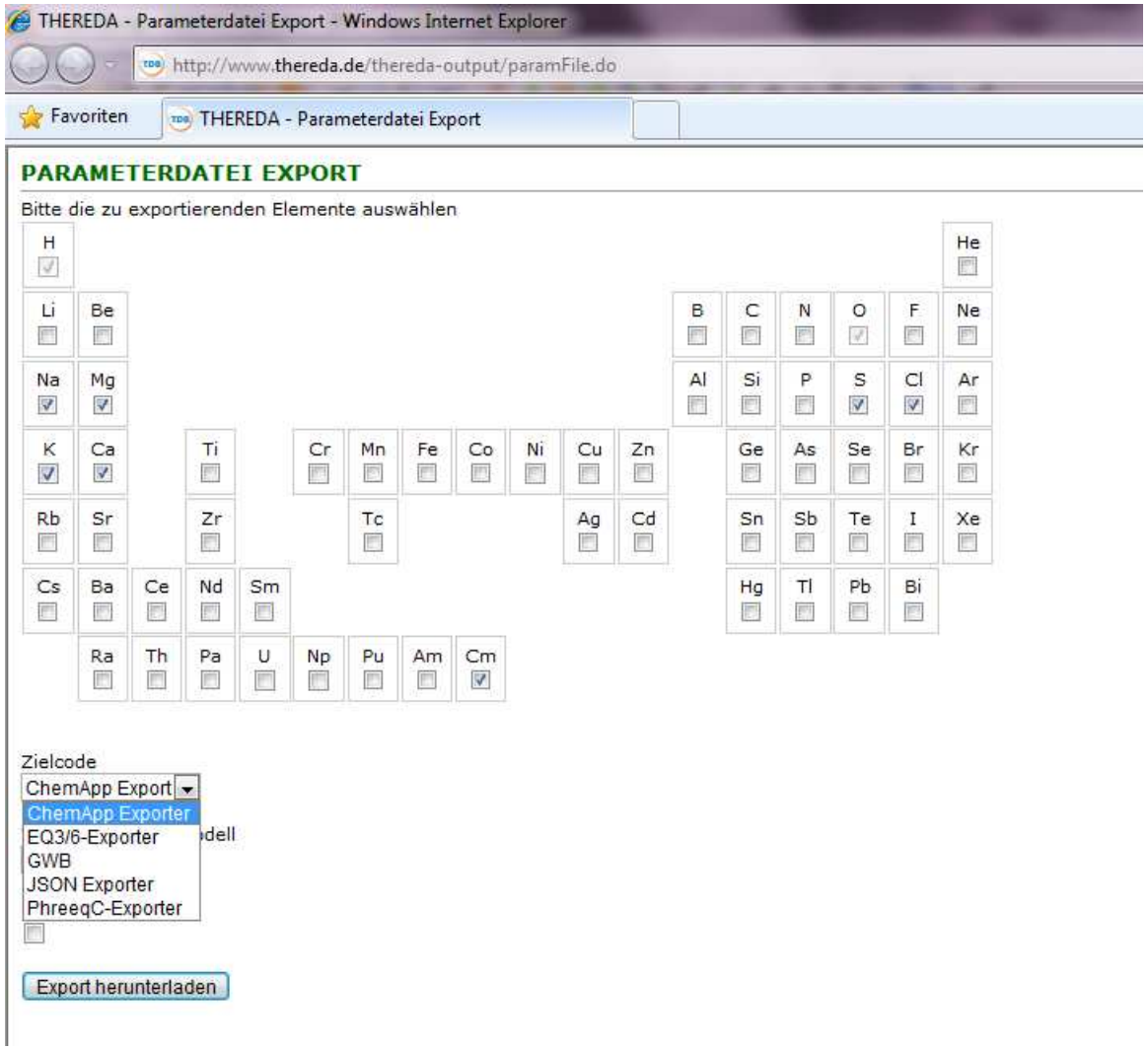
Access via World
Compilation of complex dat



Access via World Wide Web
Compilation of complex datasets (parameter files)

- Storage of md5-checksum of all downloaded parameter files
- Benchmark calculations are documented, archived and made accessible to the user

Access via World Wide Web
Compilation of complex datasets (parameter files)



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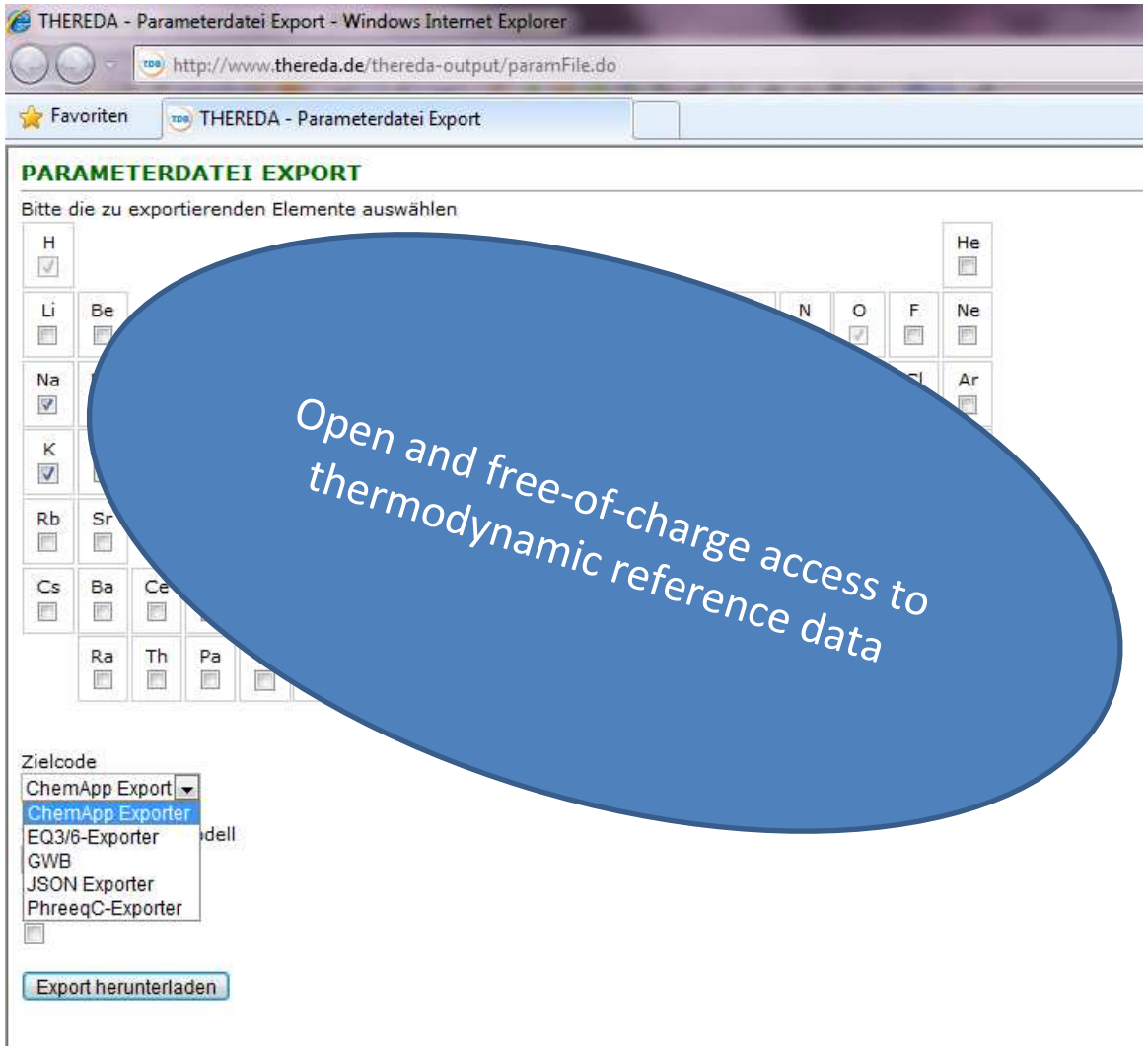
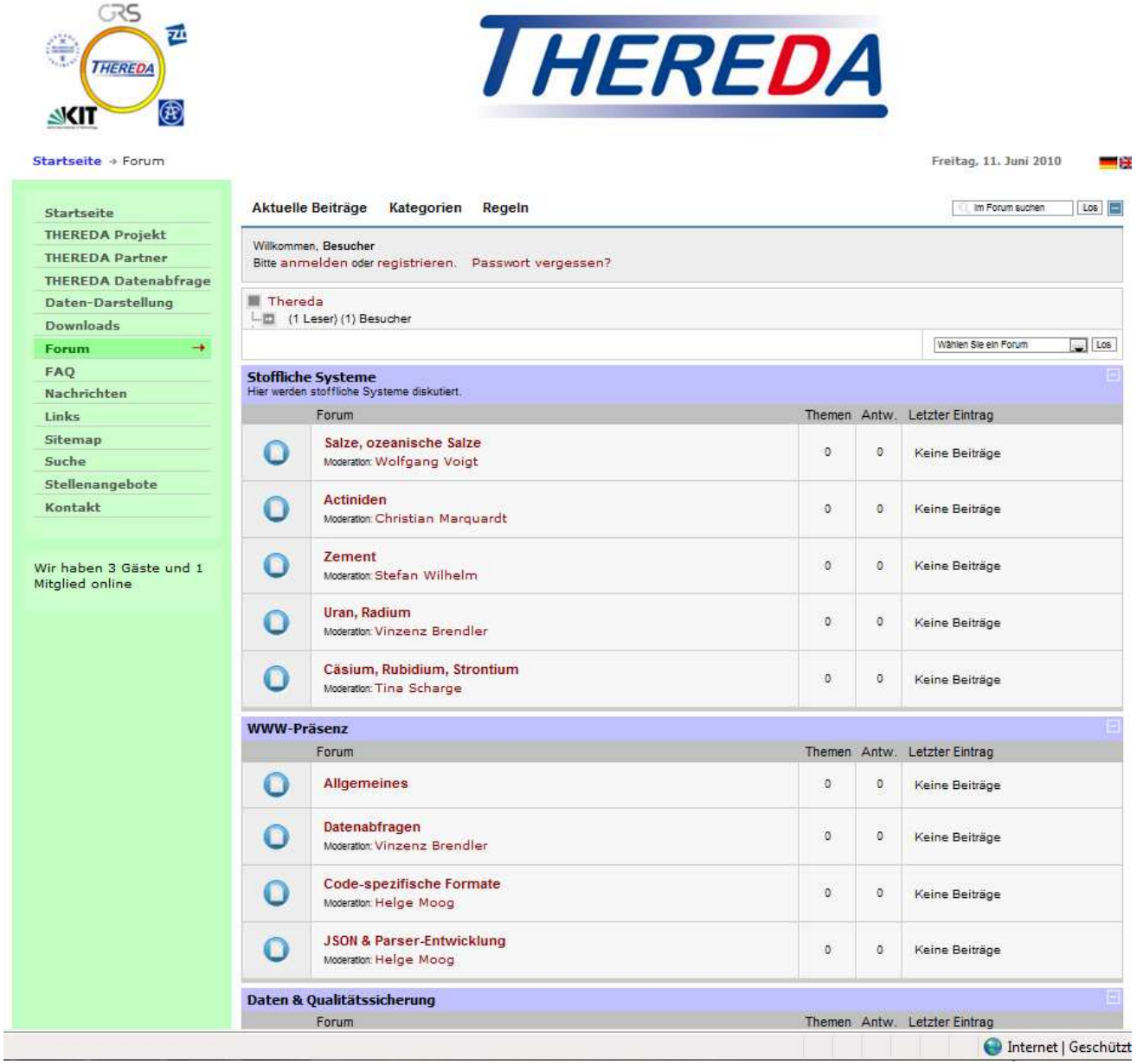
Compilation of complex datasets (parameter files)



Additional element of quality assurance: feedback from the user

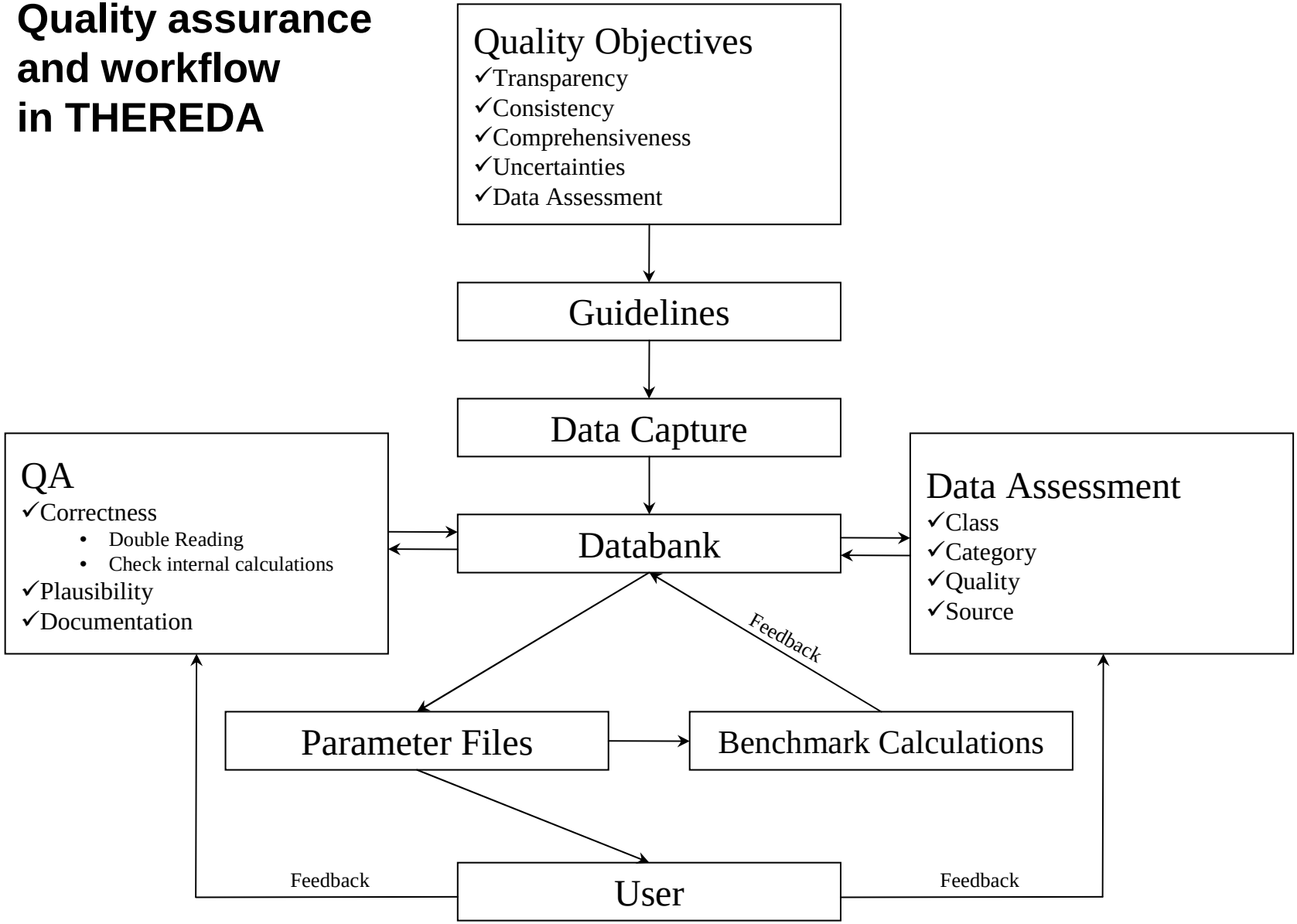


Comprehensiveness (in part)

Umfang

- 196 Phases
- 312 aq. Species (incl. 34 Primary Master)
- 336 formation reactions
- 789 Standard data sets (Pitzer)
- 717 interactions (Pitzer and SIT)

Quality assurance and workflow in THEREDA



Current activities

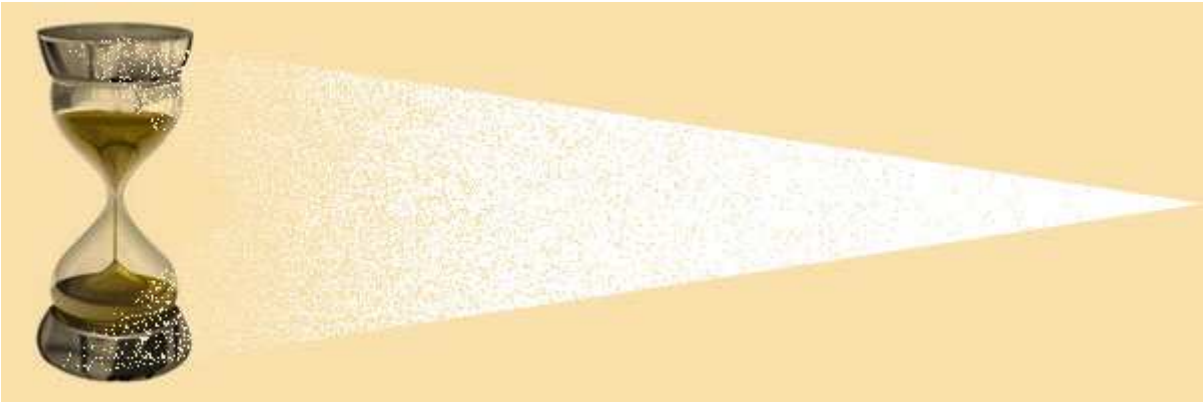
- Preparation of first release of data for
  - System of ozeanic salts (for some subsystems up to 300℃)
  - Am, Cm, Nd
- Completion (and test!) of export programs for
  - Geochemist‘s Workbench (GWB)
  - CHEMAPP
  - EQ3/6
  - PHREEQC
- Execution and documentation of benchmark calculations

Planned activities in the near future

- Planning for web-based user interface (primarly intended for editors)
- Implementation of an internal review (to be tested with the first data to be released)
- Preparation of the integration of sorption data
- Release of data for Np und Pu planned ~ Sommer 2011

Long-term useability

- Useage of open-source programs
- Low degree of abstraction of the data model
- Documention of databank structure
- Flexible databank structure
- Joint project of five research institutions



Conclusion

- THEREDA offers a web-based, state-of-the-art thermodynamic reference database for aqeous systems in equilibrium with nuclear or non-nuclear waste forms in Germany
- Joint-project of five research institutions
- Comprehensive range of data
- Access to code-specific parameter files
- Means of directing future research and of quality assurance for gouvernment agencies, servide provider, and research institutions
- Future extensions of the thermodynamic database for nuclear and non-nuclear waste forms in Gemany in conjunction with THEREDA

Vielen Dank für Ihre Aufmerksamkeit!