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European Partnership
on Radioactive Waste Management

THE NEED FOR THERMODYNAMIC DATA AND MODELING FROM THE PERSPECTIVE OF APPLICATION-ORIENTED ACTIVITIES ONGOING IN TASK 5 IN RAMPEC

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EURAD ANNUAL EVENT, DITUSC + RAMPEC JOINT SESSION

Wednesday 10th September • JC Robinet (Andra)

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INTRODUCTION

Benchmark activities within Task 5 will determine the status of the modelling capabilities for representing the effects of perturbation on radionuclide migration **at the repository cell scale**.

- Perturbation can enhance the mobility of radionuclides and increase the dose impact
- Modelling the effects of perturbations is crucial for safety assessments of all repository types

The benchmark exercise aims to compare the results of various approaches, analyse how they handle the same issues, and identify knowledge gaps in link with :

- Experimental data (RAMPEC Task 3 + SOTA),
- Macro-scale modelling (RAMPEC Task 4 + SOTA)
- **Thermodynamic data - DITUSC.**

RAMPEC TASK 5 BENCHMARK

The benchmark will be based on generic but realistic cases.

→The systems will be simplified to ease comparisons but not too much to keep the objective of evaluating the modelling potentials with regard to several phenomenological processes

→Benchmark analysis will focus on the modelling strategy as well as the quantitative results

A Post-closure Safety orientated benchmark

- Geometries of the waste cell and waste packaging;
- The properties of the materials (lining, packaging…) and the host-rock;
- The properties of the waste (type of salts, type of organics) and inventory (total mass of salt…);
- Different types of radionuclides.

Andra – CIEMAT – ASNR – LEI – EDF – UOrléans – NRG

BENCHMARK DESCRIPTION

Evaluation of the effect of a chemical perturbation generated by the waste on radionuclide's migration at the cell scale

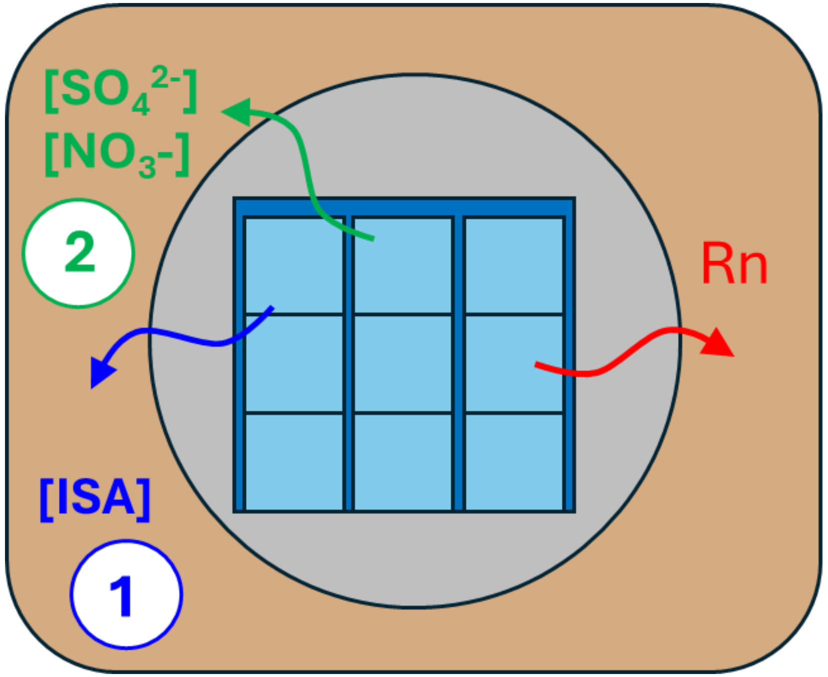
Main characteristics

- Geometry: Cylindrical waste cell (10 m in diameter) with cement lining in a compact clay-rock

- Waste
  - 1. Saline waste (nitrate/sulfate)
  - 2. organic waste (cellulosic material) → ISA

Output: (radial) chemical plume (nitrate/ sulfates or ISA) and the effect on radionuclides affected by the perturbations (Ni, Pu....)

→ Time – space evolution of concentrations

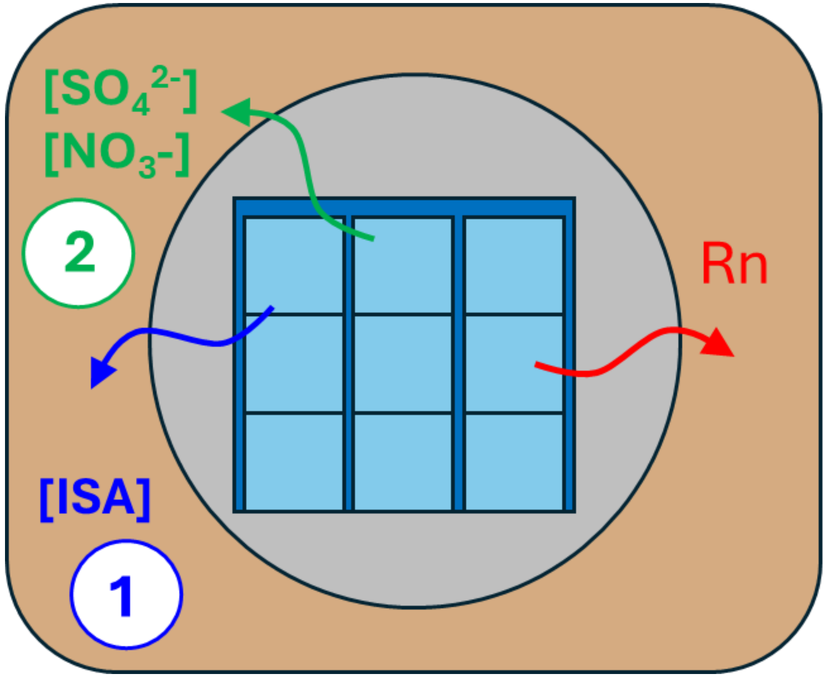


BENCHMARK DESCRIPTION

Evaluation of the effect of a chemical perturbation generated by the waste on radionuclide's migration at the cell scale

Main characteristics

- Fully saturated conditions
- Diffusion only
- Limited number of radionuclides to deal with (HTO, Cl, Ni, Ca, Am/Eu + Option : U)
- 3 cases :
  - Without pertubations
  - High and low perturbations



PHENOMENOLOGY / PROCESSES

Radionuclide migration without perturbation

- Radionuclide released from the waste
- Degradation of cement materials (with different states)
- Solubility of radionuclides
- Sorption of radionuclides onto the clay-rock and cement
    - Evolution of the cement with time
- Diffusion of radionuclides
    - Effect of electrostatic interactions in clay rock (anion exclusion/enhanced diffusion of cations)
- Evolution of the interface between cement and clay-rock
    - Evolution of RN mobility (sorption on Zeolithes ? Evolution of porosity…)

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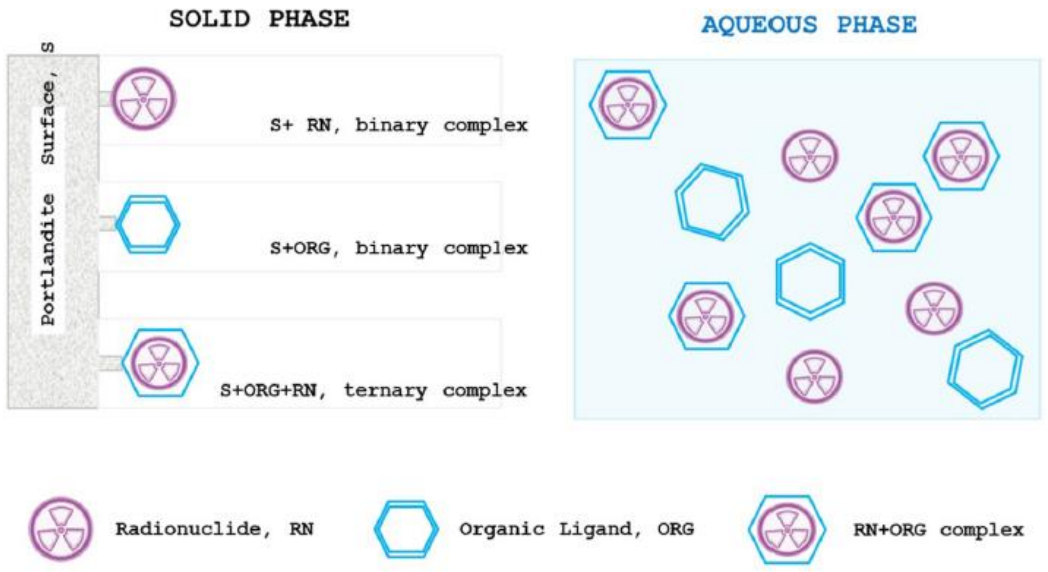
PHENOMENOLOGY / PROCESSES

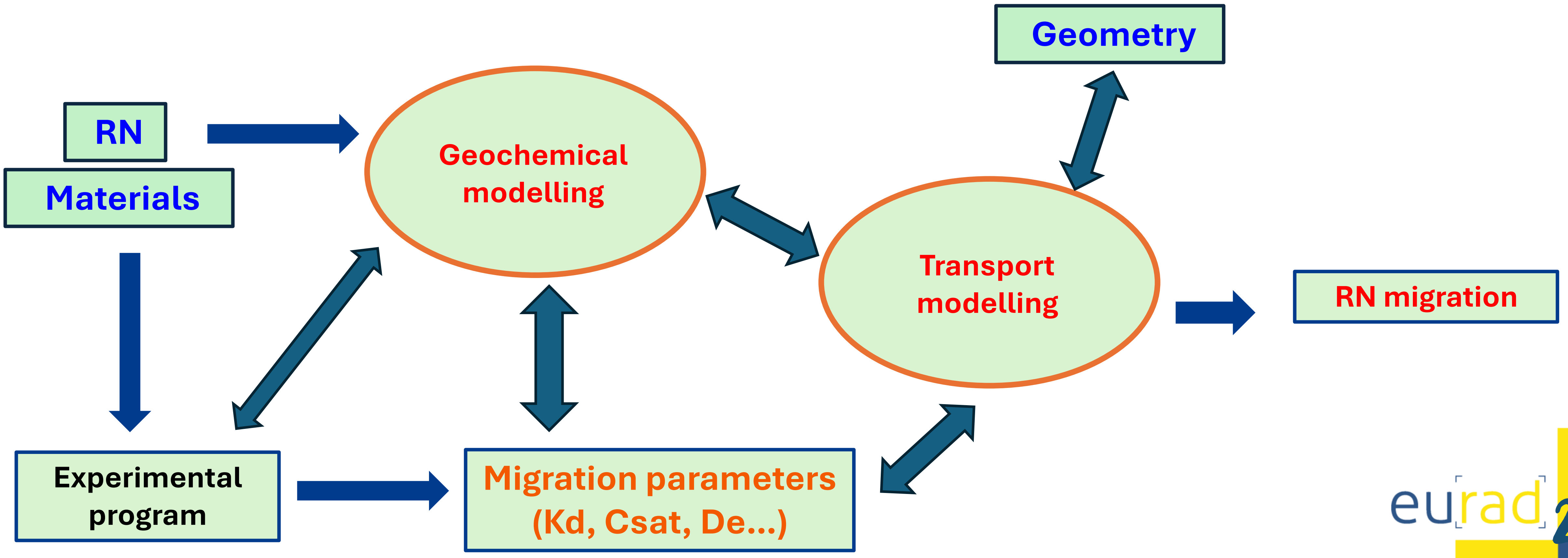
Radionuclide migration under organic or saline perturbation

- ISA/ organics and soluble salts released from the waste
- Solubility/sorption of radionuclides with ISA / Organics
- Solubility/sorption of ISA / organics
- Stability of mineral with sulfate and nitrates
- Diffusion of aqueous complexes (RN – organics)
- Aqueous speciation for high ionic strength
- Diffusion of RN under high ionic strength
- Coupled transport processes (osmotic flux)



From Almendros-Ginestà et al., 2025

MODELLING APPROACHES



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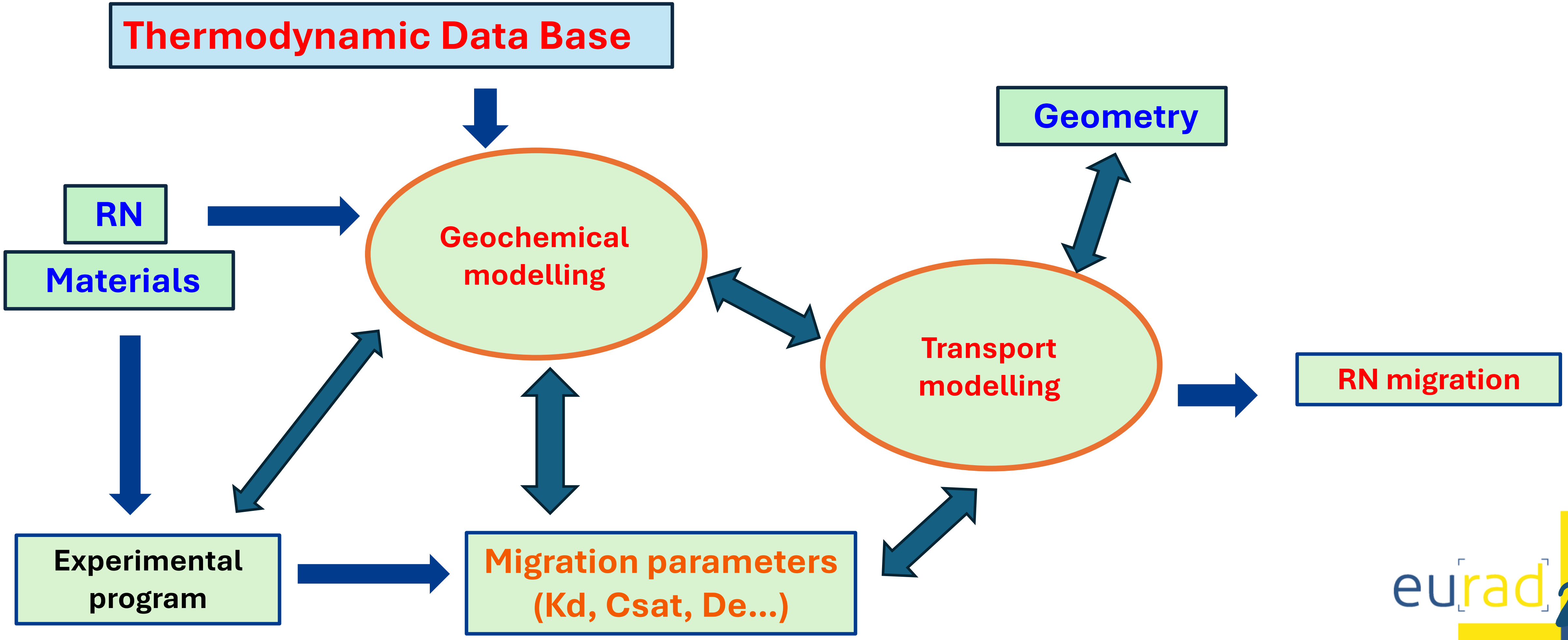
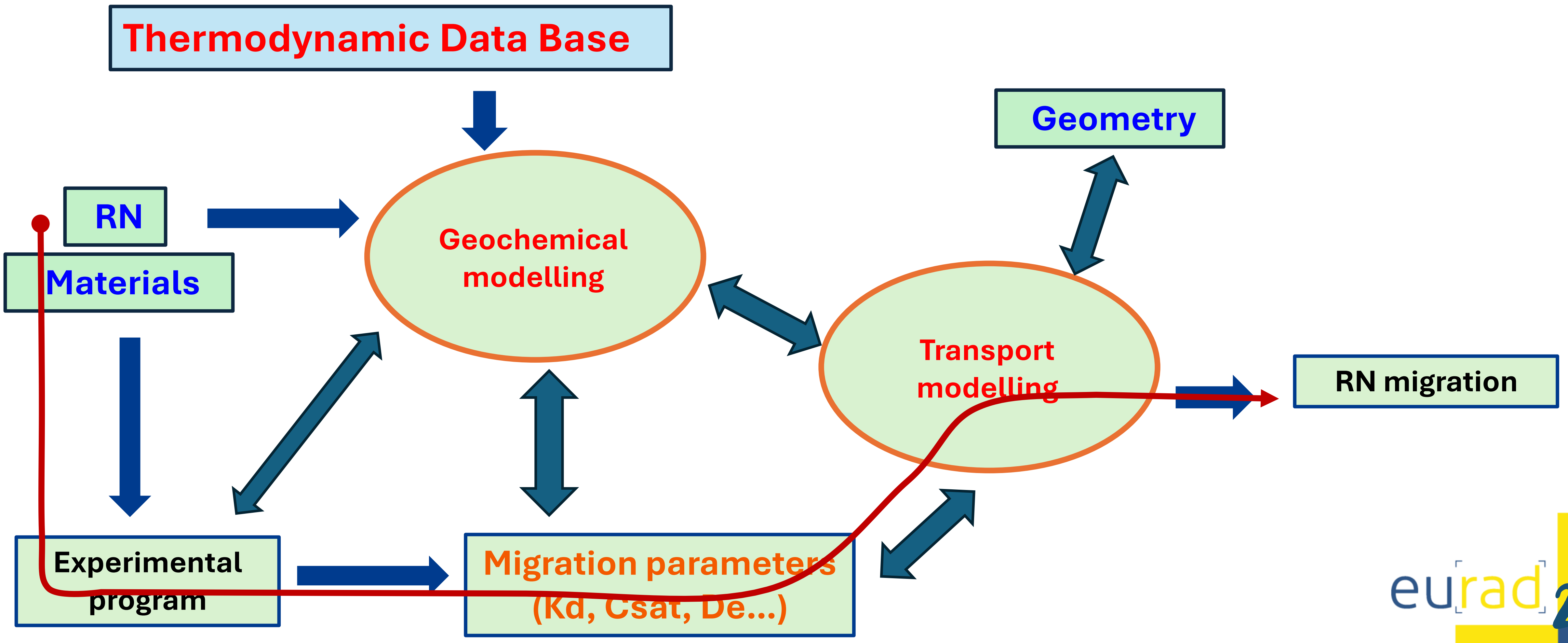
MODELLING APPROACHES



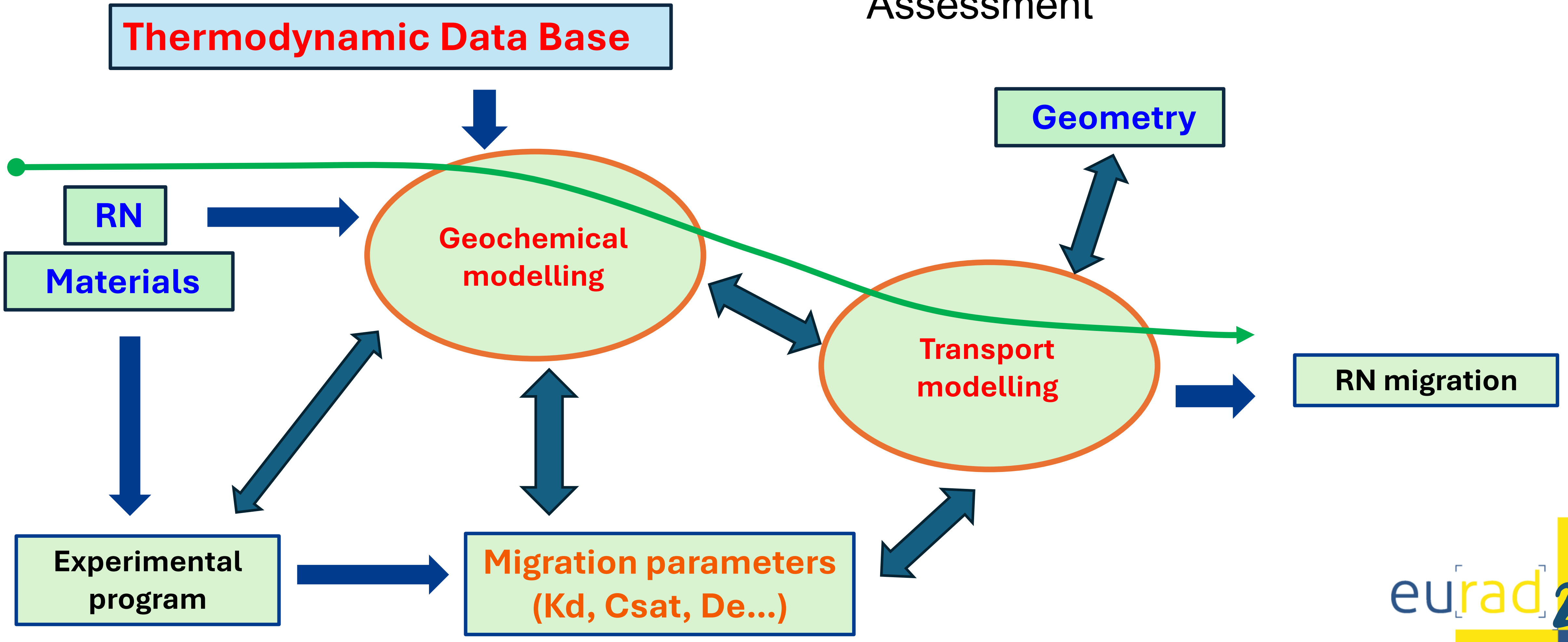
MODELLING APPROACHES

Mass transport modelling
→ mainly used for dose calculations



MODELLING APPROACHES

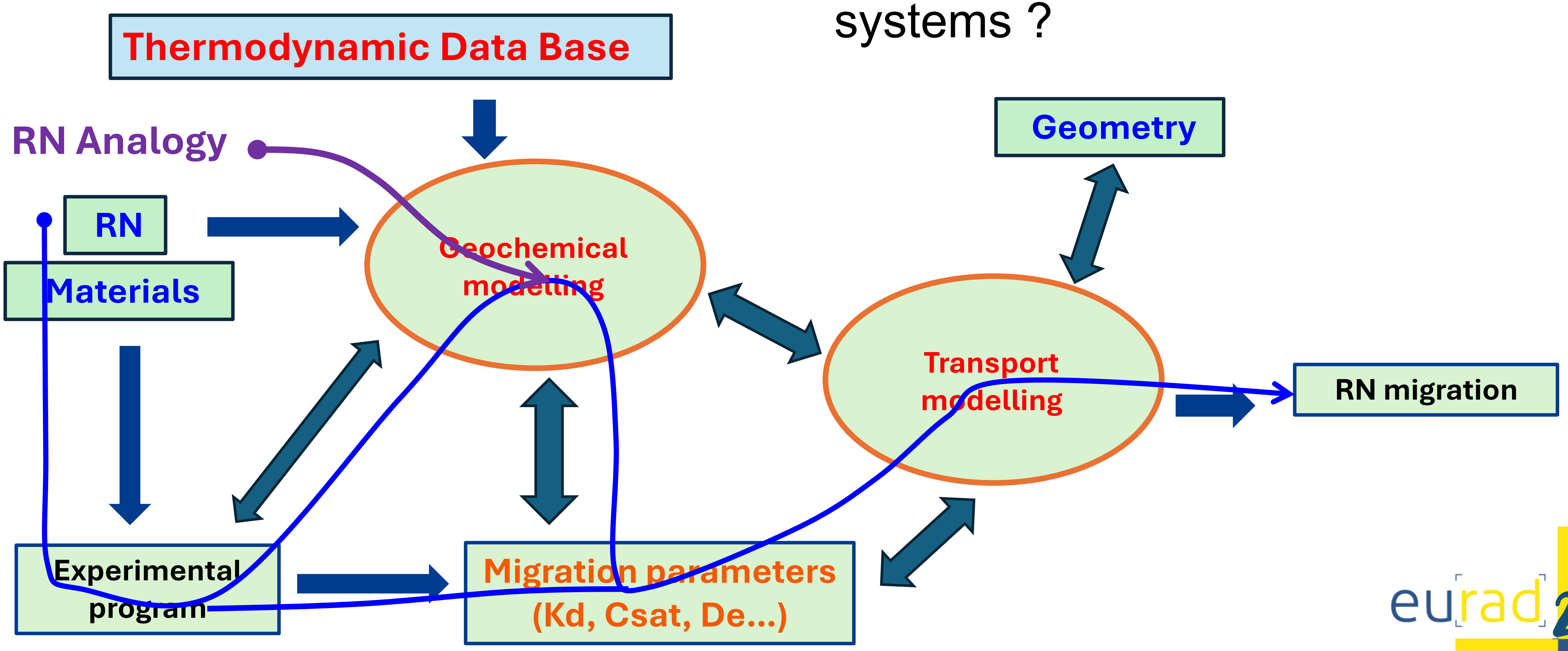
Reactive transport modelling
→ mainly used for Performance Assessment



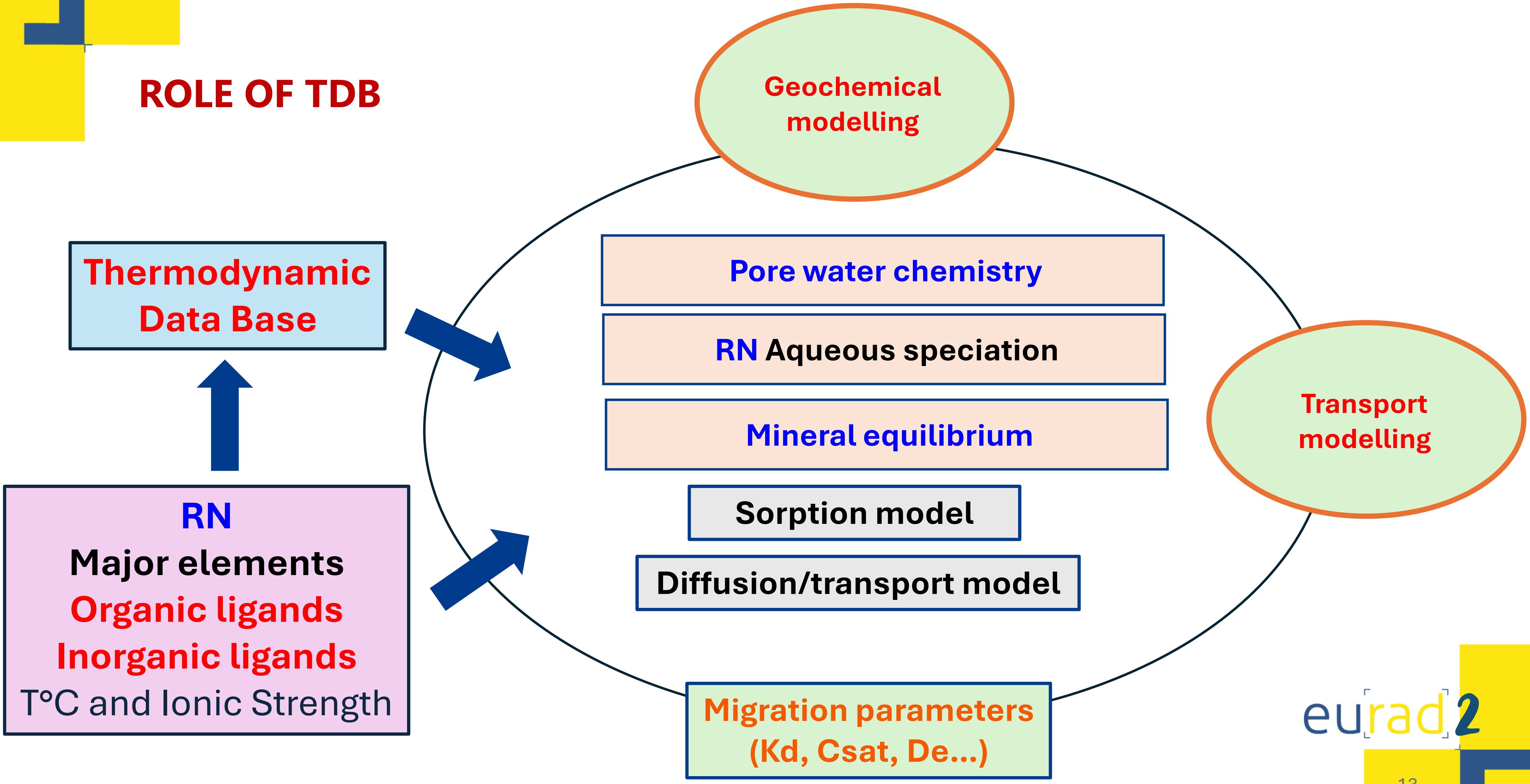
MODELLING APPROACHES

Mixed approaches

→ Alternatives to handle complex systems ?



ROLE OF TDB



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MODELLING APPROACHES

TBD is an important component of the Safety Case, even if full reactive transport modelling is not (or rarely) used to model the migration of radionuclides for dose assessment

TBD is **crucial for modelling geochemical processes** (complex geochemistry) in conjunction with transport processes and for defining parameters, but it requires a connection to a specific model (e.g. a sorption model).

To achieve this objective, **TBD must be sufficiently complete and robust** to handle the relevant processes and thus contribute efficiently to safety assessment.

→ **Identification and reduction of TDB gaps contribute to enhance the capability to manage the effect of pertubations in SA**

TDB KNOWLEDGE GAPS TOWARD RADIONUCLIDE MIGRATION

- Aqueous speciation of radionuclides : lots of data with specific issues : redox control, RN complexes (ex. RN-Ca/Mg-CO_3)
- TDB sorption model in clay-rocks : several models developed with a good constancy with experimental data – uniformity of models ? heavy to handle if there is no chemical evolution – sorption of RN complexes ?
- Generic TDB sorption model in cement considering their evolution : more complex than for clay today not at the same level→ limitations of the modelling capability ?
- Evolution of the interface between cement and clay-rock : more data needed on specific minerals (zeolithes), link between thermodynamic approaches and kinetics, sorption of RN on zeolithes …

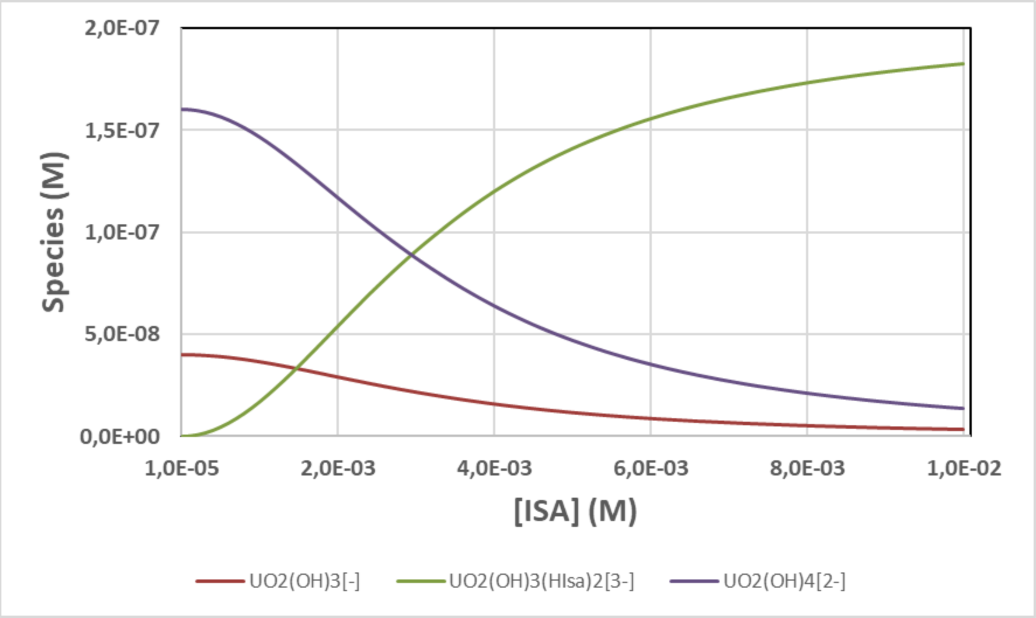
→ *How to simplify the cement-clay issue to model RN migration ?*

TDB KNOWLEDGE GAPS TOWARD RN MIGRATION UNDER PERTURBATION

Can we predict Rn behavior in the presence of ISA exiting thermodynamic data ?

List of ISA complexes in Thermochimie TDB (v12)

Species	Species	Species
Am(OH)2(HIsa)	Mg(HIsa)+	Pu(OH)4(HIsa)2-2
Ca(HIsa)+	Mg(OH)(HIsa)	Pu(OH)5(HIsa)-2
Ca(HIsa)2(cr)	Mg(OH)2(HIsa)-	Th(OH)3(HIsa)2-
Ca(Isa)	Ni(OH)(HIsa)	Th(OH)4(HIsa)-
CaPu(OH)4(HIsa)+	Ni(OH)2(HIsa)-	Th(OH)4(HIsa)2-2
CaPu(OH)5(HIsa)	Ni(OH)3(HIsa)-2	U(OH)4(HIsa)-
Cm(OH)2(HIsa)	Np(OH)4(HIsa)-	U(OH)4(HIsa)2-2
Eu(OH)2(HIsa)	Np(OH)4(HIsa)2-2	UO2(HIsa)+
H2Isa	Pu(OH)2(HIsa)	UO2(HIsa)2
HIsa-	Pu(OH)4(HIsa)-	UO2(HIsa)3-
		UO2(OH)3(HIsa)2-3

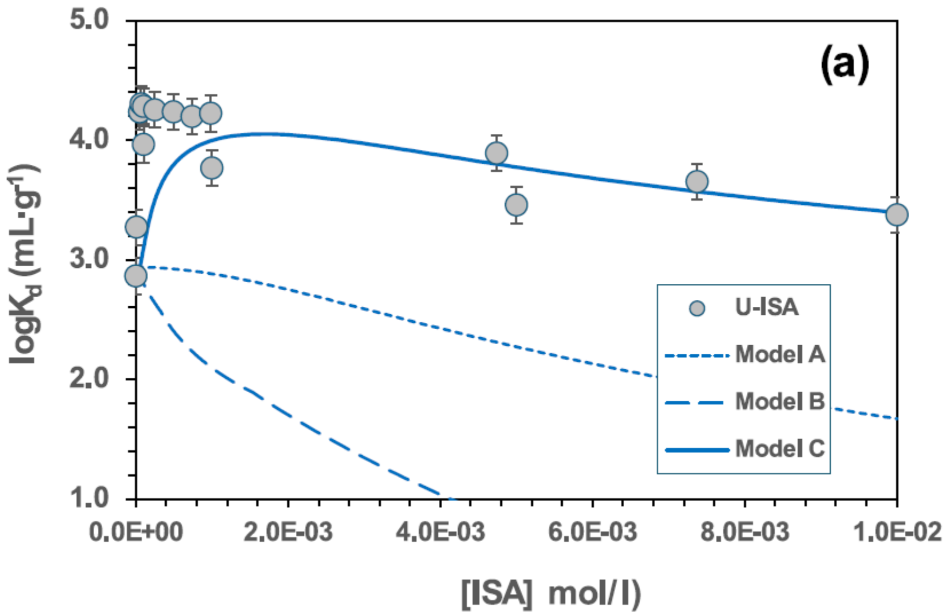


Speciation of U with ISA in cement water

From Almendros-Ginestà et al., 2025

TDB GAPS TOWARD RN MIGRATION UNDER PERTURBATION

Can we predict the behaviour of Rn in the presence of ISA using existing thermodynamic data?



Sorption of U with ISA with cement

From Almendros-Ginestà et al., 2025

Yes but…

- not with aqueous speciation only (model A)
- sorption of ISA-U complexes is needed (model C)

ISA is the best-documented organic molecule in TDB – <u>other molecules</u> ?

TDB GAPS TOWARD RADIONUCLIDE MIGRATION UNDER PERTURBATION

- Organic :
  - Aqueous speciation : many data notably for ISA but gaps identified :
    - Cations (Ca, Mg...) /RN/organic
    - Consistency between data ex. Am-ISA – Eu-ISA (cf. work of Dagnelie)
    - Lack of data for many organic molecules (PFAS, phathlates...)
  - Data on RN-Organic sorption
- Saline : data with identified gaps
  - Solubility of mixed sulfate/nitrate systems
  - Lack of data for complexes RN-NO3-, RN-SO42-
  - Use of LFER/analogies to cover data gaps?
  - Interaction SIT parameters? Pitzer approaches ?