ONDRAF/NIRAS

Some concluding remarks

R. Gens, M. Van Geet / Dpt. LT RD&D

Belgian Agency for Radioactive Waste and Enriched Fissile Materials

Spent Fuel

- **Spent fuel behaviour & dissolution rate**
  - Key step: U IV → U VI
  - Oxidative dissolution driven by radiolysis
    - Same radiolytic yields at neutral and high pH
  - Very similar behaviour in cementitious environment and neutral/slightly alkaline media (in terms of dissolution rates) ⇒ we can rely on existing databases
  - Data coherent with other programs
  - Alpha threshold is environment dependent
    - No Eh poising capacity associated to the concrete buffer(CEM I)

ONDRAF/NIRAS

Spent Fuel

- **H_2 inhibits UO_2 dissolution (very low residual rates)**
  - Detailed mechanisms still raising questions

- **In presence of hydrogen and/or below alpha threshold**
  - Dissolution mainly controlled by chemical equilibriums (solubility, diffusion/sorption)
    - SA: not a key parameter for non oxidative dissolution
    - Lifetime: millions of years

ONDRAF/NIRAS

Spent Fuel

- **Future program will focus on the following topics**
  - Need additional evidences, datasets for confidence building (robustness)
    - New pgm with real spent fuels under preparation in close collaboration with and between KIT & SCK●CEN
  - Decrease uncertainty on alpha threshold in cementitious environment (→ related to overpack lifetime & early canister failure scenario)
  - Tests at low hydrogen partial pressures (passive corrosion of steel overpack & insert)
    - Data needed for alpha doped materials (with ε particles?)
  - IRF
    - Few data for high burn up UOX and MOX fuels
      - Follow up of international & national programs
      - Participation to EC First Nuclides Project

ONDRAF/NIRAS

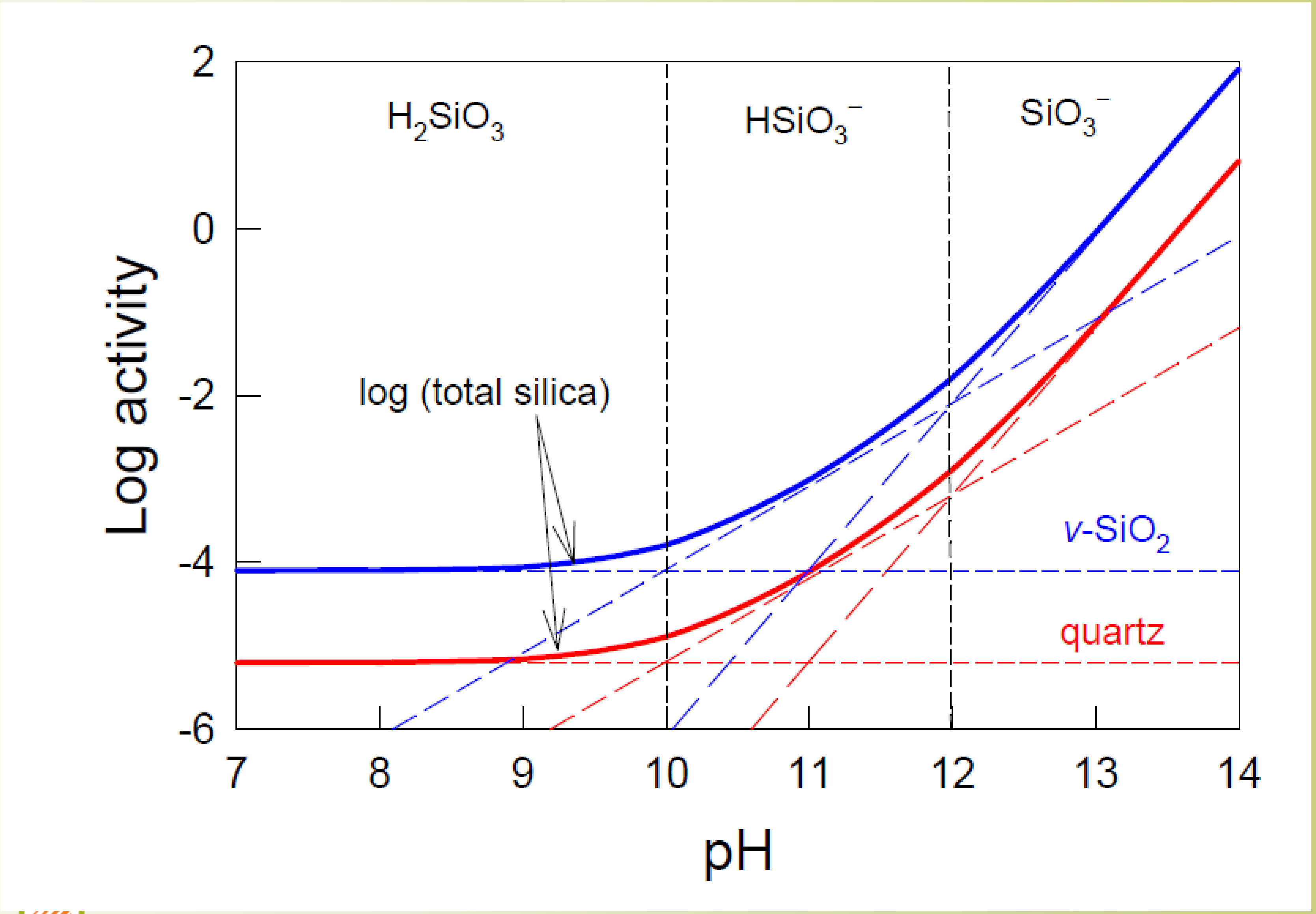
Glass

- **Same fundamental dissolution mechanisms as in neutral – slightly alkaline media**
  - But different kinetic regimes/controlling steps
  - High initial dissolution rate

- **At high pH: main dissolution drivers**
  - Attack of silica network (Si-O-Si bridges broken)
    - Formation of A-S-H/C-S-H
  - Resumption of alteration as a result of secondary phase formation (zeolites)

ONDRAF/NIRAS

Glass



ONDRAF/NIRAS

(A. Paul, 1990)

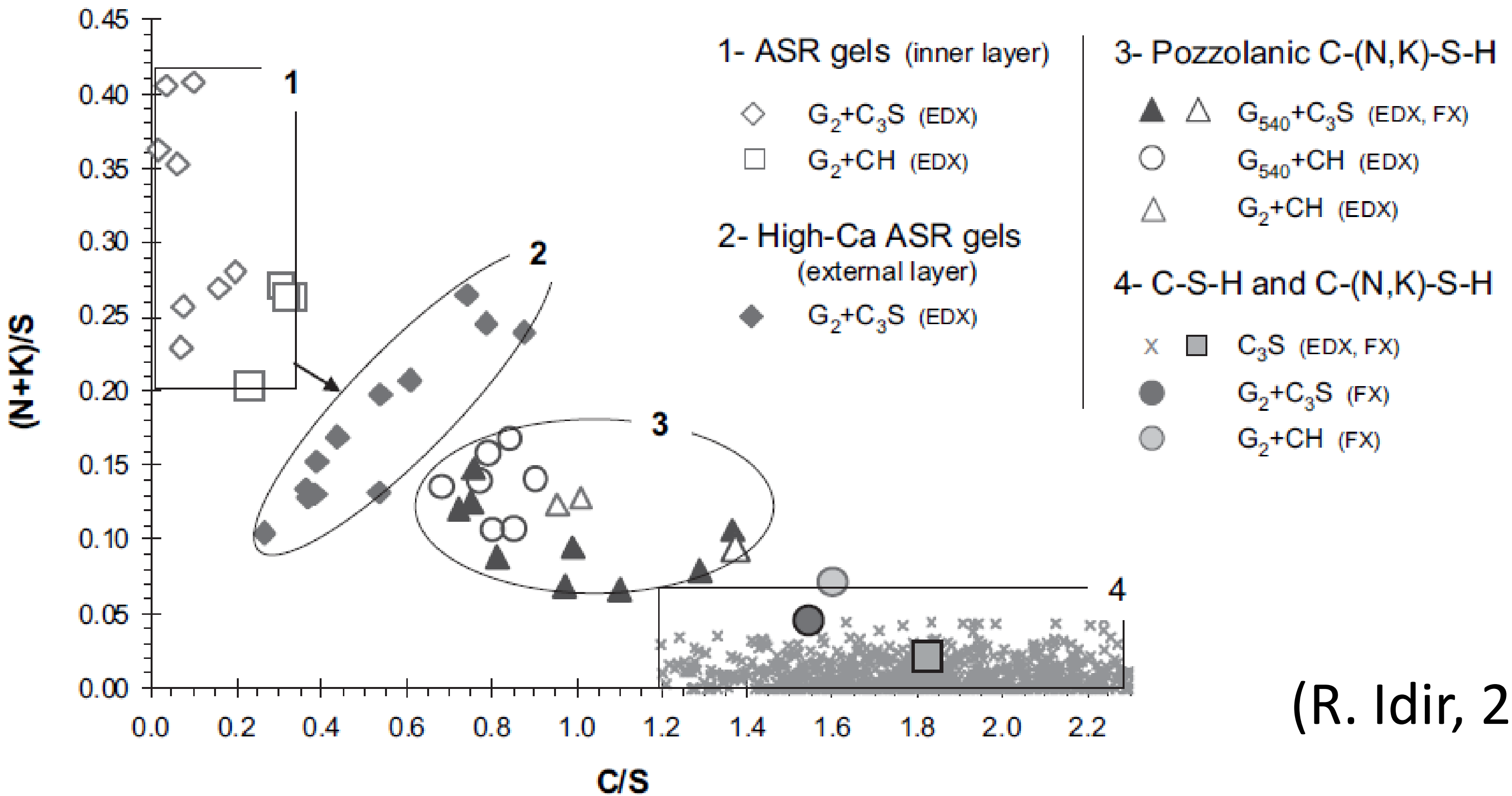
Glass

- **Cement industry: many relevant publications (different goals)**
  - R&D on ASR mechanisms (glass used as reactive "aggregate")
  - Main potential application: as SCM in concrete (ASR ⇔ *Pozzolanic reaction*)

- **SCM (BFS, FA, glass) & Radioactive vitrified waste**
  - Similar reaction path (to some extent)

ONDRAF/NIRAS

Glass



(R. Idir, 2011)

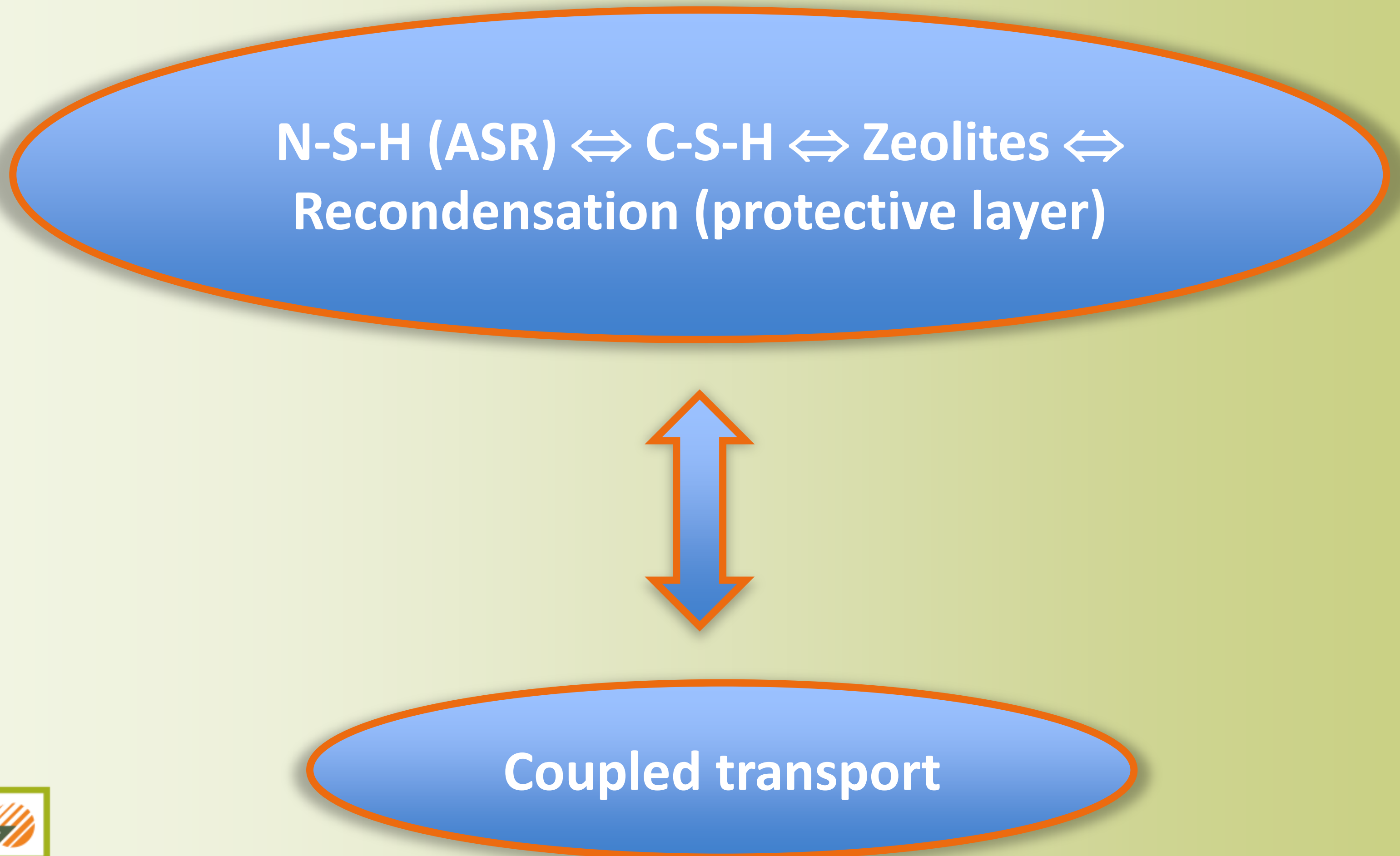
Reaction path in line with Ichikawa mechanistic model

(T. Ichikawa, 2007)

ONDRAF/NIRAS

Glass

- **Future R&D: long-term dissolution rate evolution**



ONDRAF/NIRAS

Glass

- **Experimental data and expected prevailing repository conditions (confined conditions, no renewal) suggest that the dissolution rate should decrease**
  - Confined tests (SCK●CEN): lower rates
  - Glass rods in mortar: lower ASR rate for uncracked samples (Song, 2008)
  - Already observed with Al metal in cement ("quasi" passivation)
  - Transport constrains (diffusion controlled)
    - Percolation tests (SCK●CEN): $V_{Fin} << V_{int}$
- **Filler material in the overpack: reactive silica to fix the alkalis** (prevent ASR, R. Takata, 2004)
- **Overpack lifetime > concrete stage 1 (alkalis leached out?)**

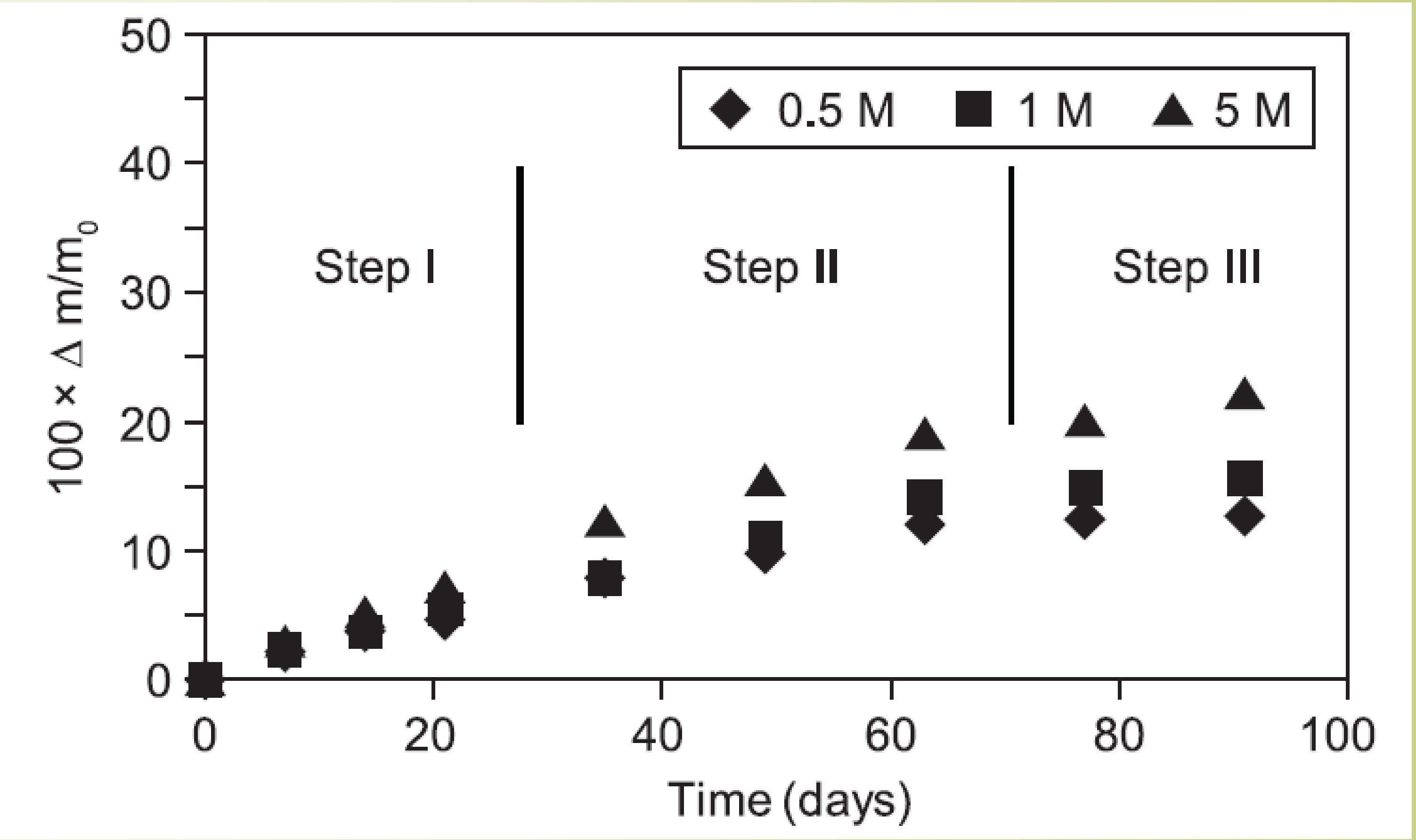
ONDRAF/NIRAS

Glass

- **Formation of a protective layer on the long term (C-S-H and/or zeolites) ?**
  - Clogging of the fractures (decrease of SA)?
  - “Cementation” observed with glass powder (SCK●CEN, CEA)
  - Dense zeolite phases observed (CEA)
- **Experimental and modeling pgm needed**
- **Objectives: to provide multiple lines of evidence (V_{LT} << V_{int})**
  - No use in PA for SFC 1
  - Safety reserve
    - Secondary phase formation: complex processes depending on many parameters (M. Fournier *et al*, 2014)
      - Kinetic & TDB: data missing
    - Multiple interfaces + evolving conditions
      - Concrete buffer/Filler/Overpack/Filler(?)/Canister/glass

ONDRAF/NIRAS

Glass



(S. Kouassi *et al.*, 2010)

ONDRAF/NIRAS

Glass

- **Concrete ⇔ Clay**
  - No ideal design optimizing the durability and performance of all EBS components
- **Clay**
  - LT tests (SCK●CEN): no leveling off after 10 years
  - Fe-Glass interaction: could keep high initial dissolution rate during some (long?) transient (G. de Combarieu *et al*, 2011)
- **Confinement phase (overpack + glass)**
  - SAFIR 2 (clay): ~ 75 000 years
  - Supercontainer (concrete): ~ 100 000 years (**at least**)

ONDRAF/NIRAS

General conclusions

- **10 years ago: change of design (Supercontainer, concrete buffer)**
  - Challenging situation: only very few relevant data available (very limited interest in other countries)
- **10 years later: significant progress + international collaboration**
  - Significant progress in the development of the scientific assessment basis (databases, mechanistic understanding)
    - Spent fuel: bridges between Belgian and other national pgm (no different behaviour in concrete)
      - R&D performed by KIT & SCK
      - Expert Panel review (on an yearly basis)
    - Glass
      - Studies performed in France (CSD-B) and UK (Magnox glass)
      - Scientific collaboration with AREVA, CEA, ANDRA, Subatech

ONDRAF/NIRAS

General conclusions

- **Coralus**
  - Highly valuable technical &scientific outcomes
  - REX: relevant input for potential future *in situ* pgm (Supercontainer Design)

- **Future R&D**

  - Knowledge gaps and priorities well identified (see RD&D plan)
  - Will take place in the frame of international collaborations

ONDRAF/NIRAS