Karlsruhe Institute of Technology

Institute for Transuranium Elements

Verhalten von Actiniden unter endlagerrelevanten Bedingungen

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Introduction

Nuclear waste disposal in deep geological formations:

- Actinides (*Plutonium*) responsible for long-term radiotoxicity of HLW / spent fuel.
- Aquatic actinide chemistry relevant in the case of water intrusion into a repository.

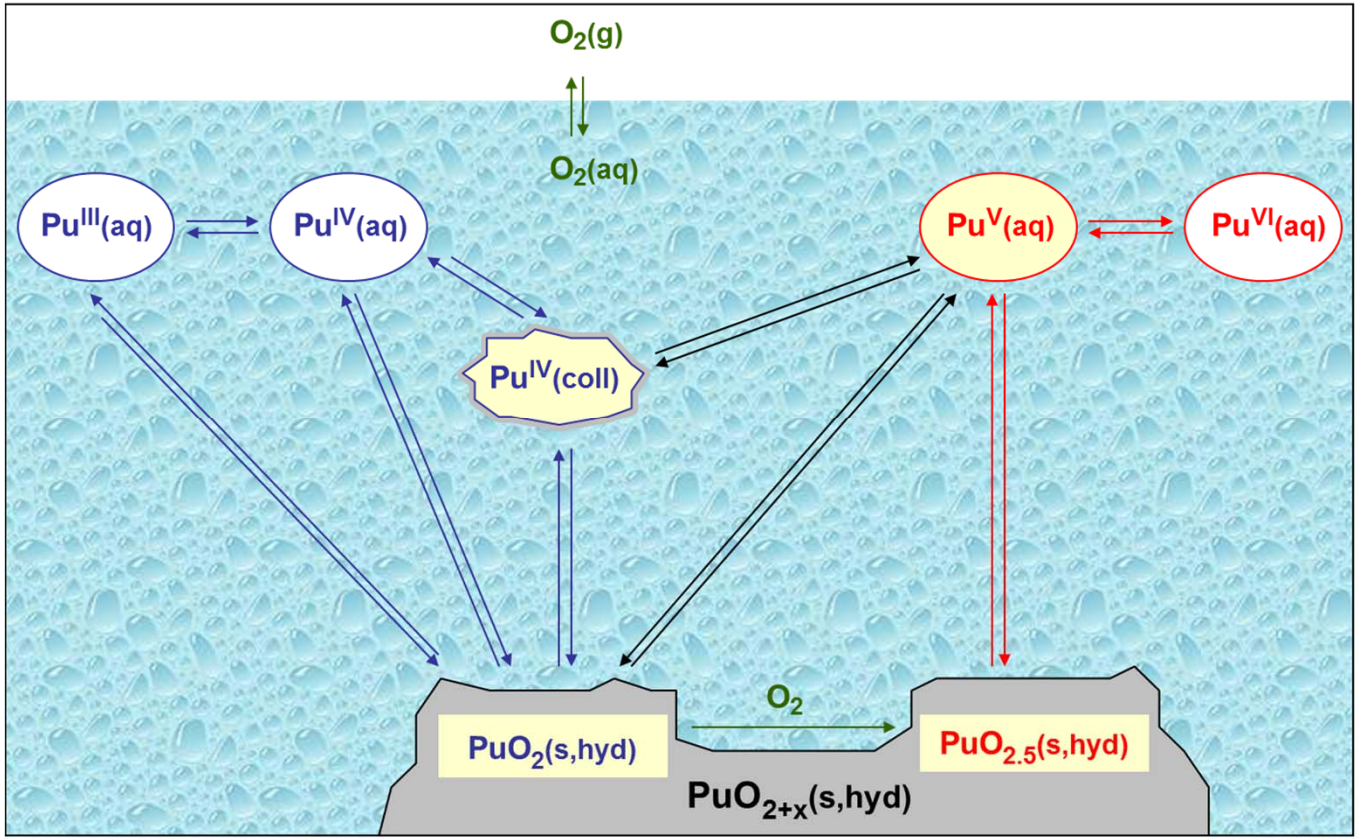
  ⇒ For reliable assessment of long-term safety of nuclear waste repositories:

  prediction of **actinide solubility and speciation** basic requirement.

Actinide redox properties

- Actinides known to exist/coexist in different oxidation states, e.g. U(IV,VI), Np(IV,V,VI), Pu(III,IV,V,VI).
- Chemical behavior depends strongly on redox state.
- Understanding of **actinide redox behavior** mandatory for prediction of actinide solubility.

Introduction



Topics of PhD work:

- **solubility of Pu(IV)** in alkaline $CaCl_2$ solutions.

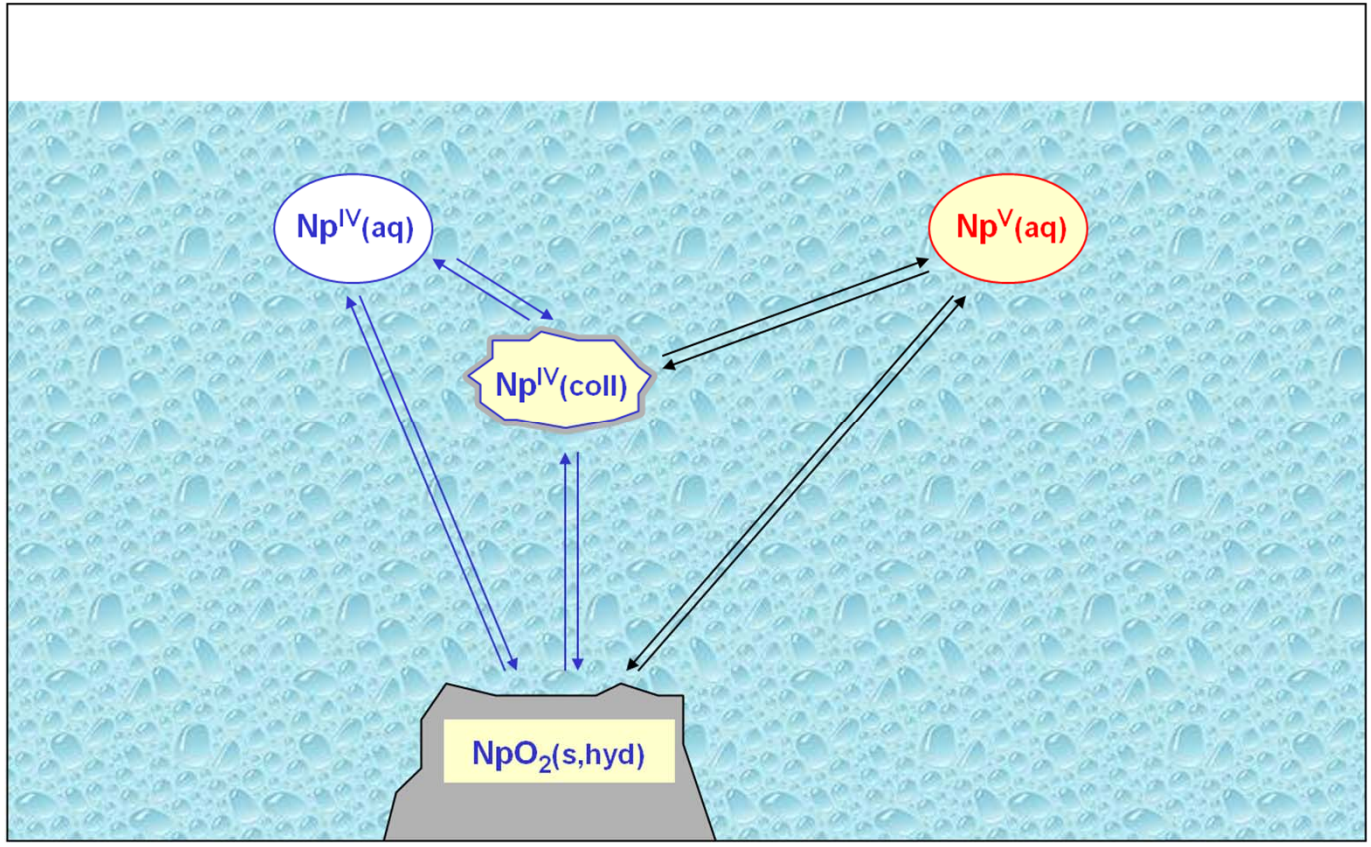
- **solubility of Pu(VI)** in alkaline $CaCl_2$ solutions.

- **Pu redox behavior** under reducing conditions: Pu(III) ⇔ Pu(IV) equilibrium

Aqueous Plutonium chemistry is complex due to:

- coexistence of up to four oxidation states,
- polymeric and colloidal species,
- complex solid phases.

Introduction



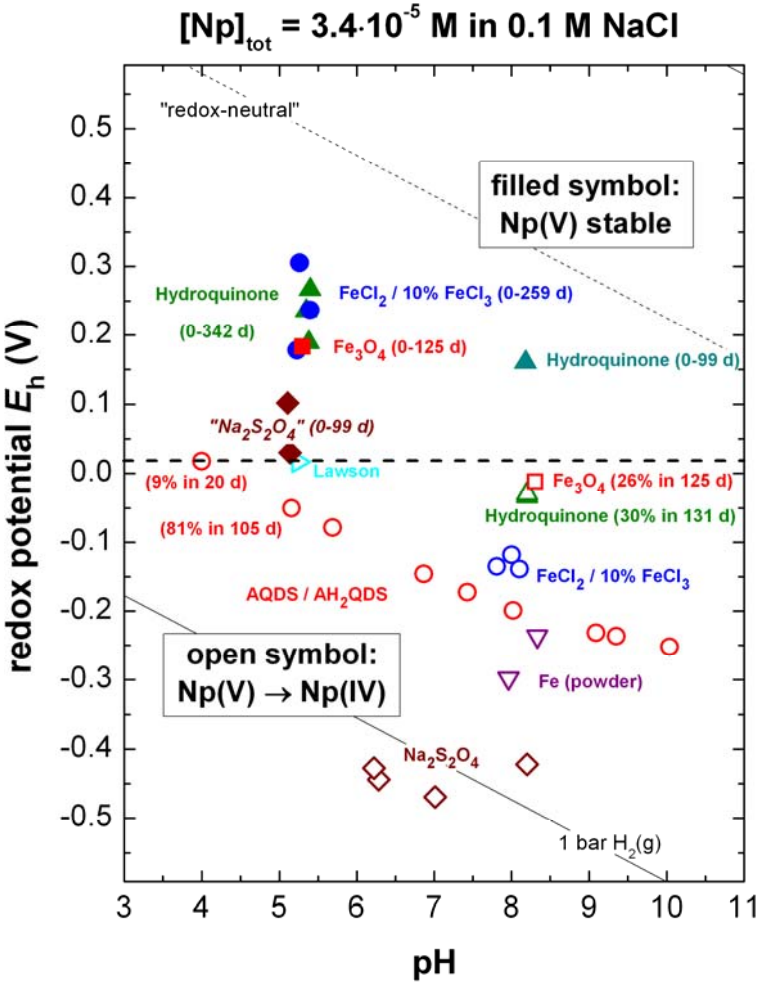
Neptunium: chemically simpler analogue

Topics of PhD work:

- **solubility of Np(IV)** in alkaline $CaCl_2$ solutions.
- **solubility of Np(V)** in alkaline $CaCl_2$ solutions.
- **solubility of Np(VI)** in alkaline $CaCl_2$ solutions.
- **Np redox behavior:** thermodynamics and kinetics of Np(V) reduction

A) Redox experiments: Np(V) → Np(IV) reduction

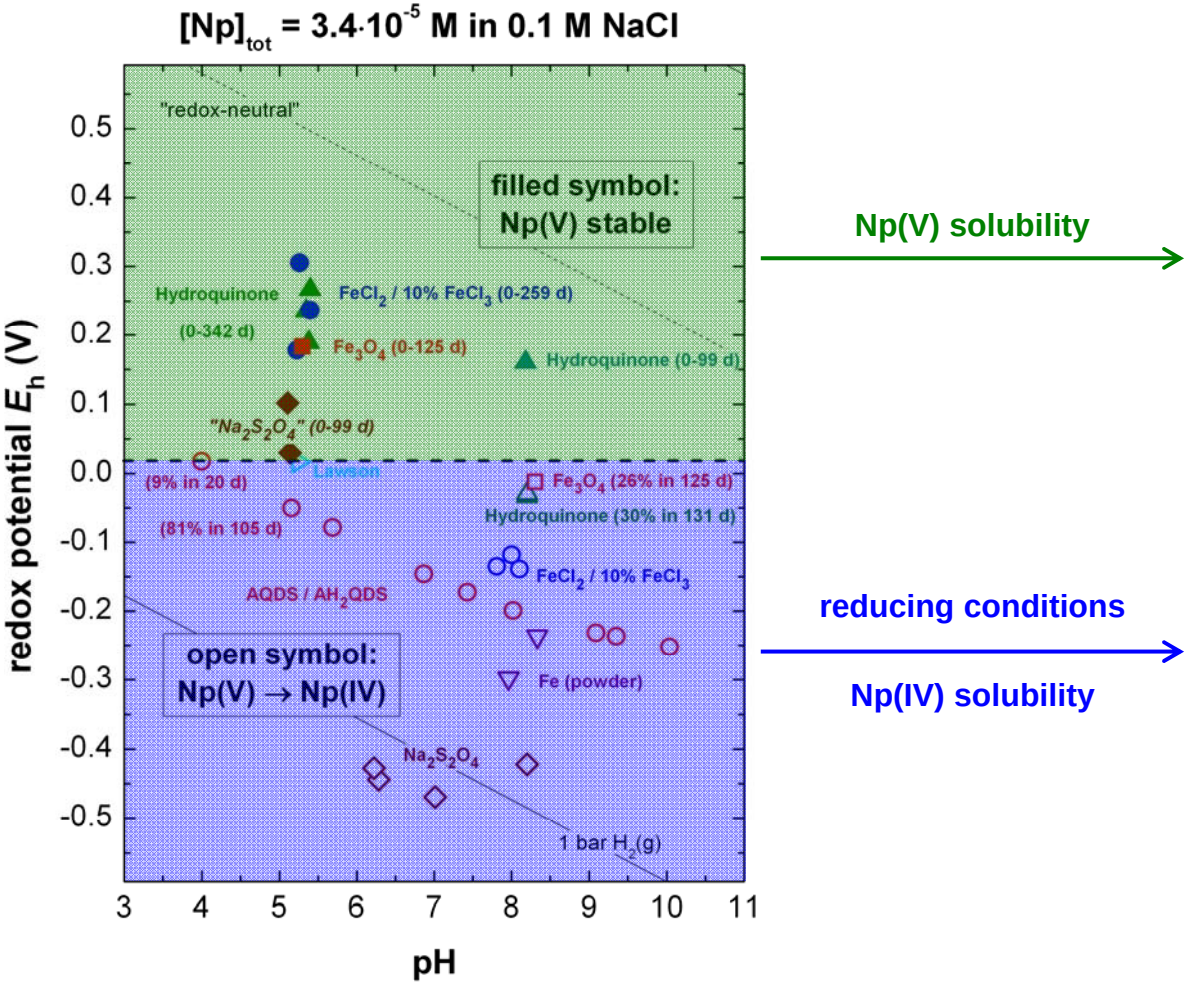
Addition of $Np(V)_{aq}$ to various reducing systems (0.1 M NaCl, pH 4 - 10): reduction YES / NO ?



B) Solubility experiments: Np in $CaCl_2$ solutions

Results of redox experiments:

E_h and pH conditions to stabilize the different Np oxidation states → solubility experiments.



Np(V) solubility

reducing conditions

Np(IV) solubility

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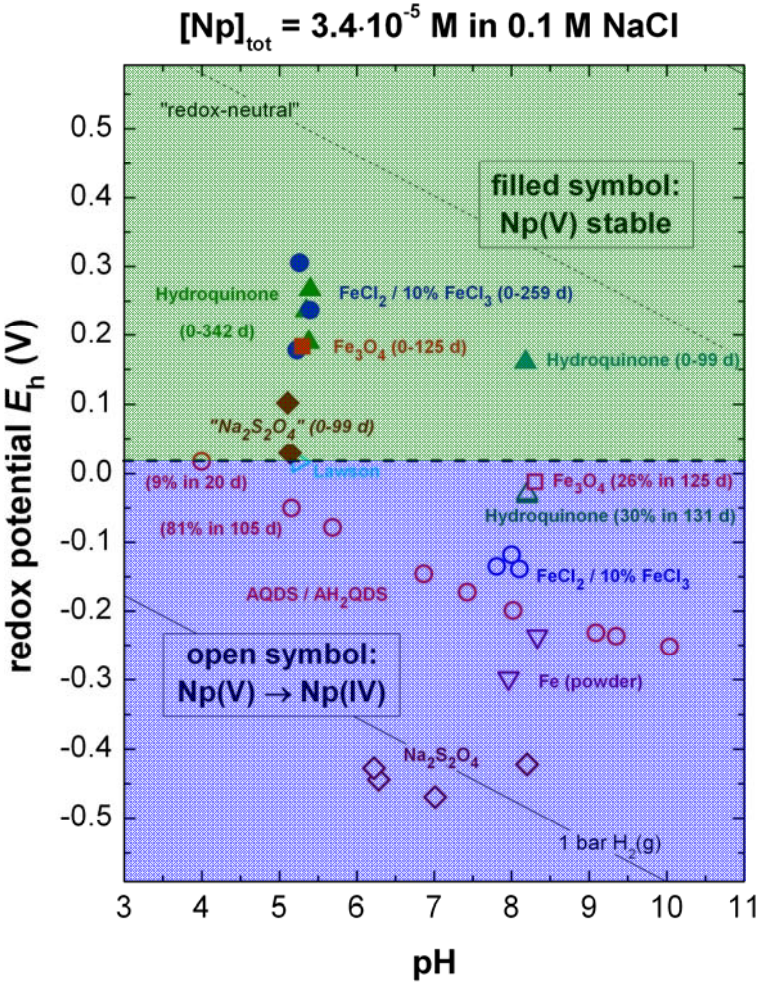

Np(V) solubility

reducing conditions

Np(IV) solubility