Investigations of the uranyl and neodymium(III) adsorption behavior on ion exchange resins for the weak-acid resin process

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Motivation

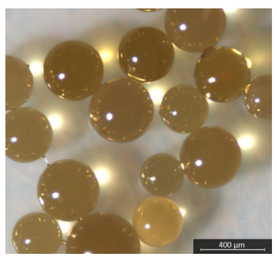
Partitioning and transmutation (P&T) is considered as one of the promising methods to safely handle minor actinides (MA's) which origin from energy production by nuclear fission. Within the ASGARD project[1], the focus is on the preparation of microspheres consisting of a homogeneous mixture of uranium and MA oxides which can be used for sphere-pac fuels[2].

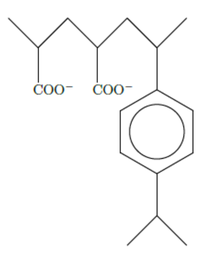
A promising method for the preparation of microspheres is the so-called weak-acid resin process[3]. In this work, the adsorption behavior of Nd^{3+} (as surrogate for trivalent actinides) and UO_2^{2+} has been investigated on the commercial weak-acid cation exchange resins Amberlite IRC-86 and Lewatit TP-207.

Weak-acid resin process

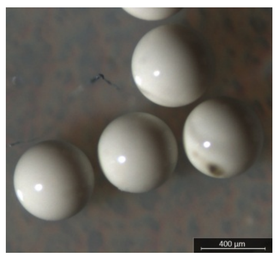
- Adsorption of UO_2^{2+} and Nd^{3+} on weak-acid ion exchange resins:
  - $zR\mathrm{COOH}\ (s) + \mathrm{Y}^{z+}\ (aq) \rightleftharpoons (R\mathrm{COO})_z\mathrm{Y}\ (s) + z\mathrm{H}^{+}\ (aq)$

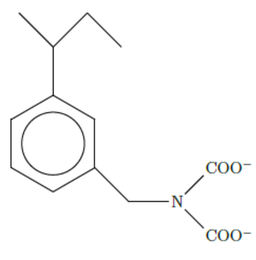
Amberlite IRC-86:
Gel-type resin
Capacity: 10.7 meq·g^{-1}
Operational pH: 5 - 10

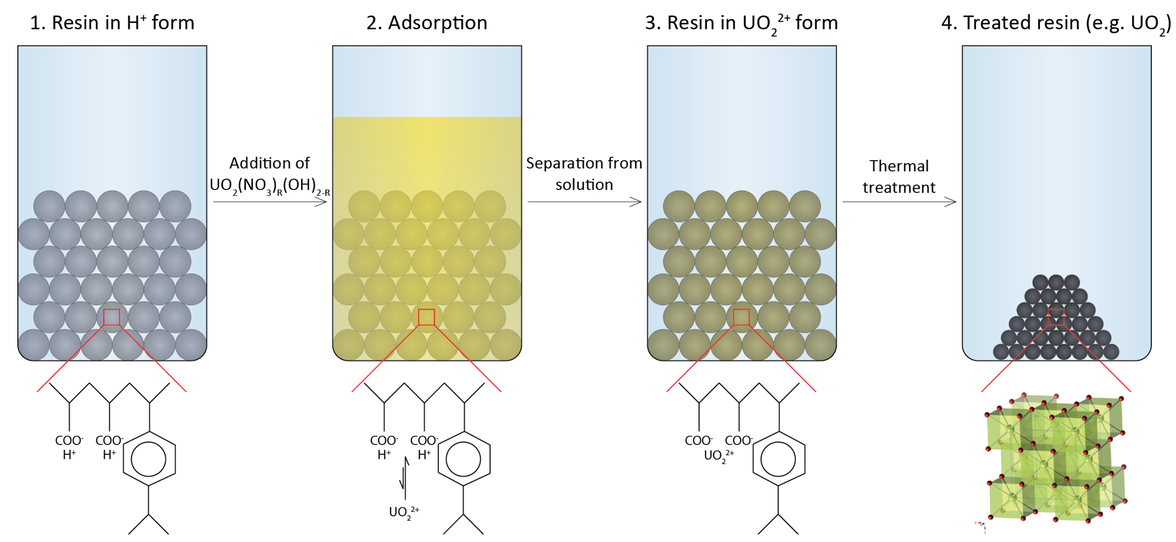




Lewatit TP-207:
Macroporous type resin
Capacity: 6.9 meq·g^{-1}
Operational pH: 1.5 - 9

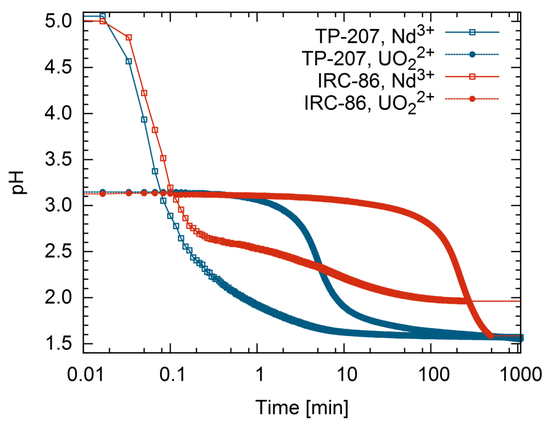


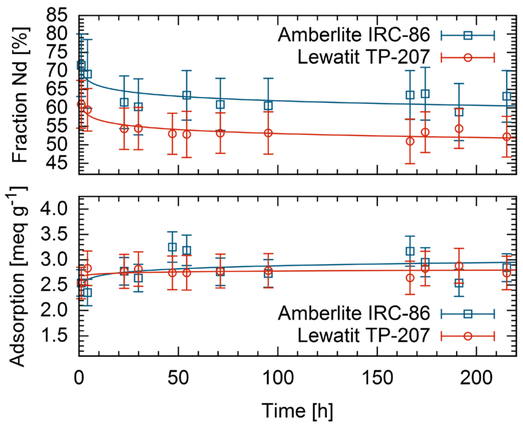


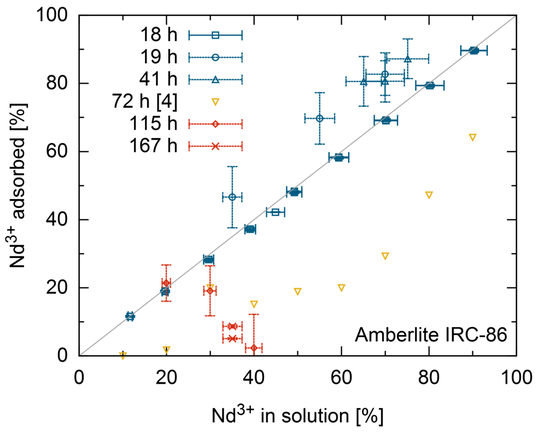


Schematic overview of the weak-acid resin process, exemplary shown for production of UO_2 microspheres using Amberlite IRC-86

Adsorption kinetics



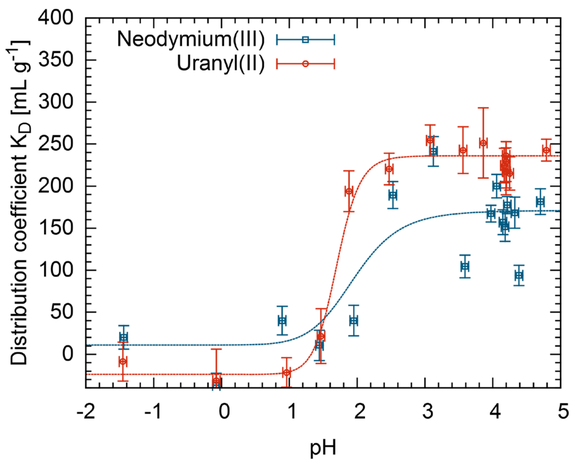




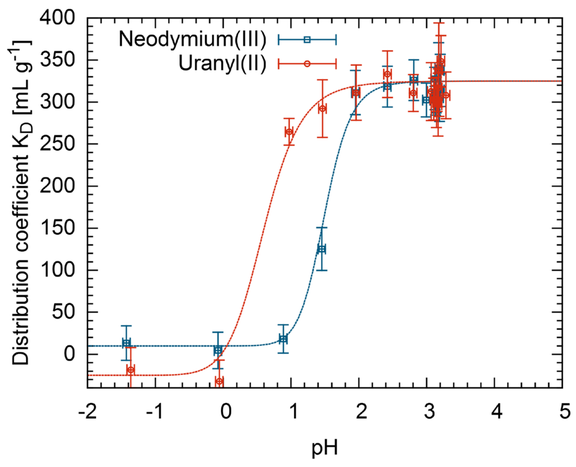
- pH decrease proportional to adsorption;
- Equilibrium after 18 h;
  - Nd^{3+} kinetically favored;
- Nd^{3+} fraction decrease with longer contact;
  - UO_2^{2+} thermodynamically favored;
- Contacting time of 18 h yield best results.

pH dependence on adsorption

Amberlite IRC-86

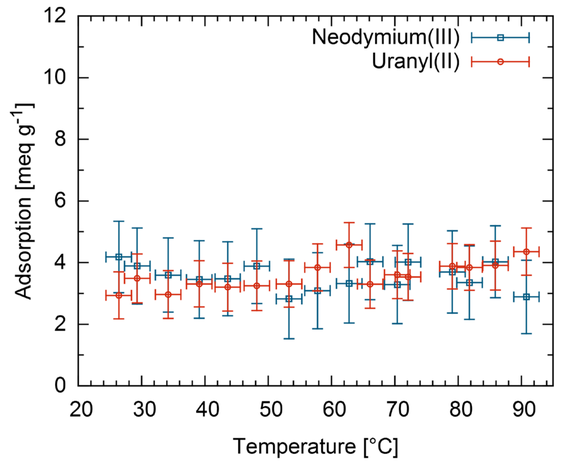


Lewatit TP-207

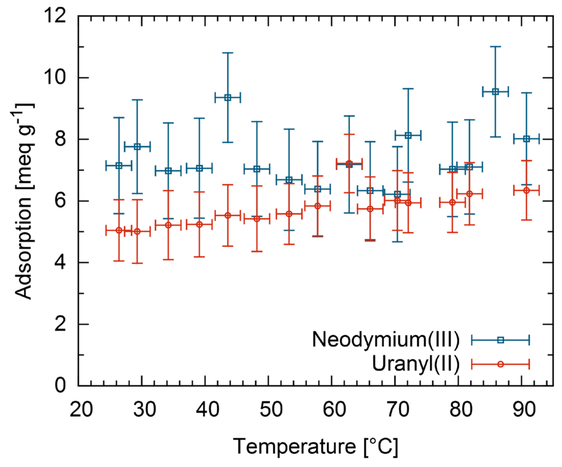


Temperature dependence on adsorption

Amberlite IRC-86

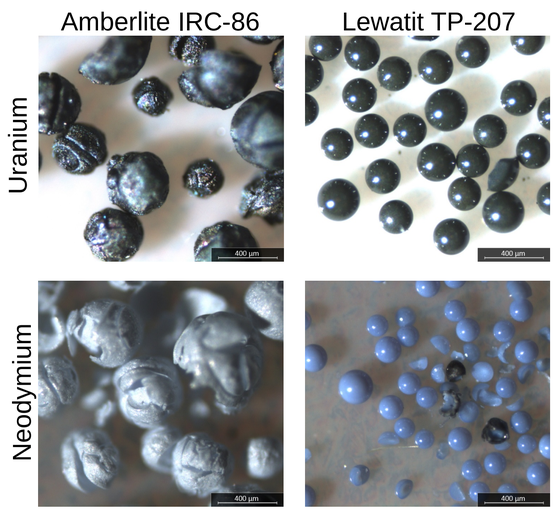


Lewatit TP-207

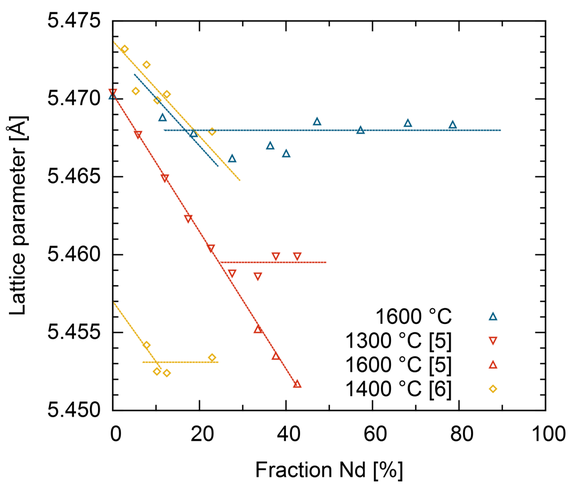


Thermal treatment

1. Oxidizing atmosphere (900 °C, Air)



2. Reducing atmosphere (1600 °C; H_2/Ar)



Conclusion

- Optimal contacting time of 18 h:
  - < 18 h: No equilibrium;
  - > 18 h: Decrease of Nd^{3+} fraction;
- Strong decrease at pH < 3;
- No significant influence of temperature;
- Loading up to 0.43 g·g^{-1} achieved;
- Amberlite IRC-86 not suitable for treatment in air;
  - Many broken particles; VIPAC possible;
- No equilibrium solid solution achieved (reducing atmosphere).

[1] Pouchon, M., *Comprehensive Nuclear Materials, 3.11 Sphere-Pac and VIPAC fuels* (2012)
[2] K. Notz et al., *Radiochimica Acta*, 25: p. 153-160 (1978).
[3] http://www.asgardproject.eu/.
[4] M. Weidenfeld, Diplomarbeit, FH Aachen University of Applied Sciences (2010).
[5] C. Schreinemachers, Master thesis, FH Aachen University of Applied Sciences (2013).
[6] L. Desgranges et al., *Inorganic Chemistry*, 51(17): p. 9147-9149 (2012).

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- All uncertainties are given as expanded uncertainties with a coverage factor *k* = 2.
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