Modelling of Am stripping step with TODGA - TPAEN

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Warsaw, April 2015

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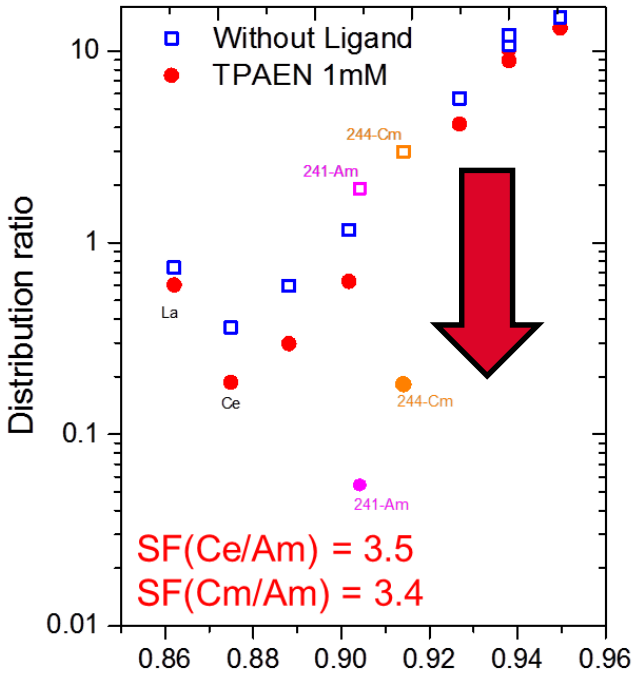
Objectives

- Discuss the feasibility of a flowsheet compared to performances, based on two approaches : theoretical stages and a first model implemented in the PAREX code
- Make a progress report on modelling and identify a list of experiments to strengthen the model
- Suggest a first flowsheet



[Ln] ~ 4 mM

Feed (mg/L)	
[La]	350
[Ce]	750
[Pr]	400
[Nd]	1400
[Am]	420

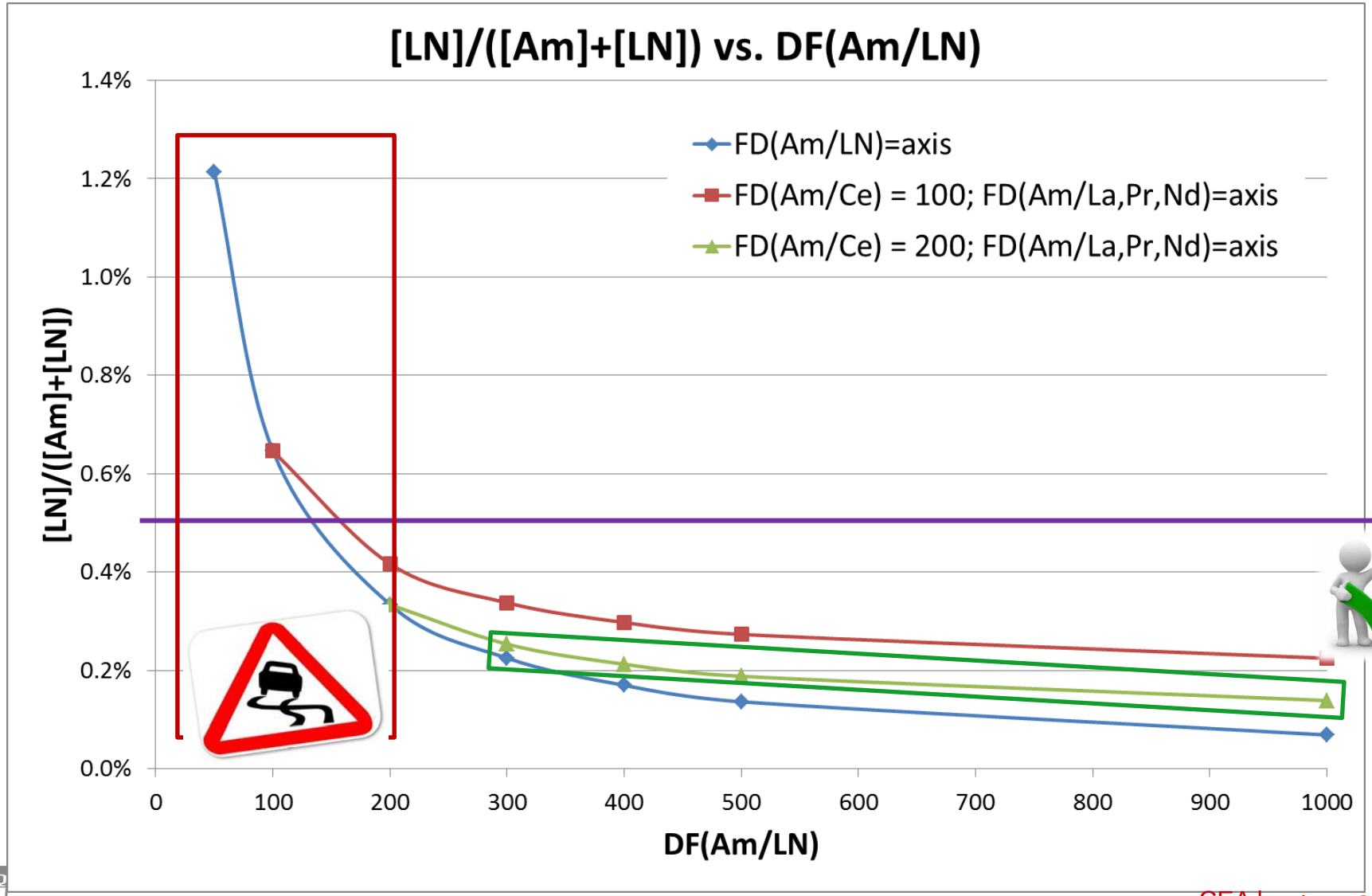
[Ln] ~ 25 mM

Target performances

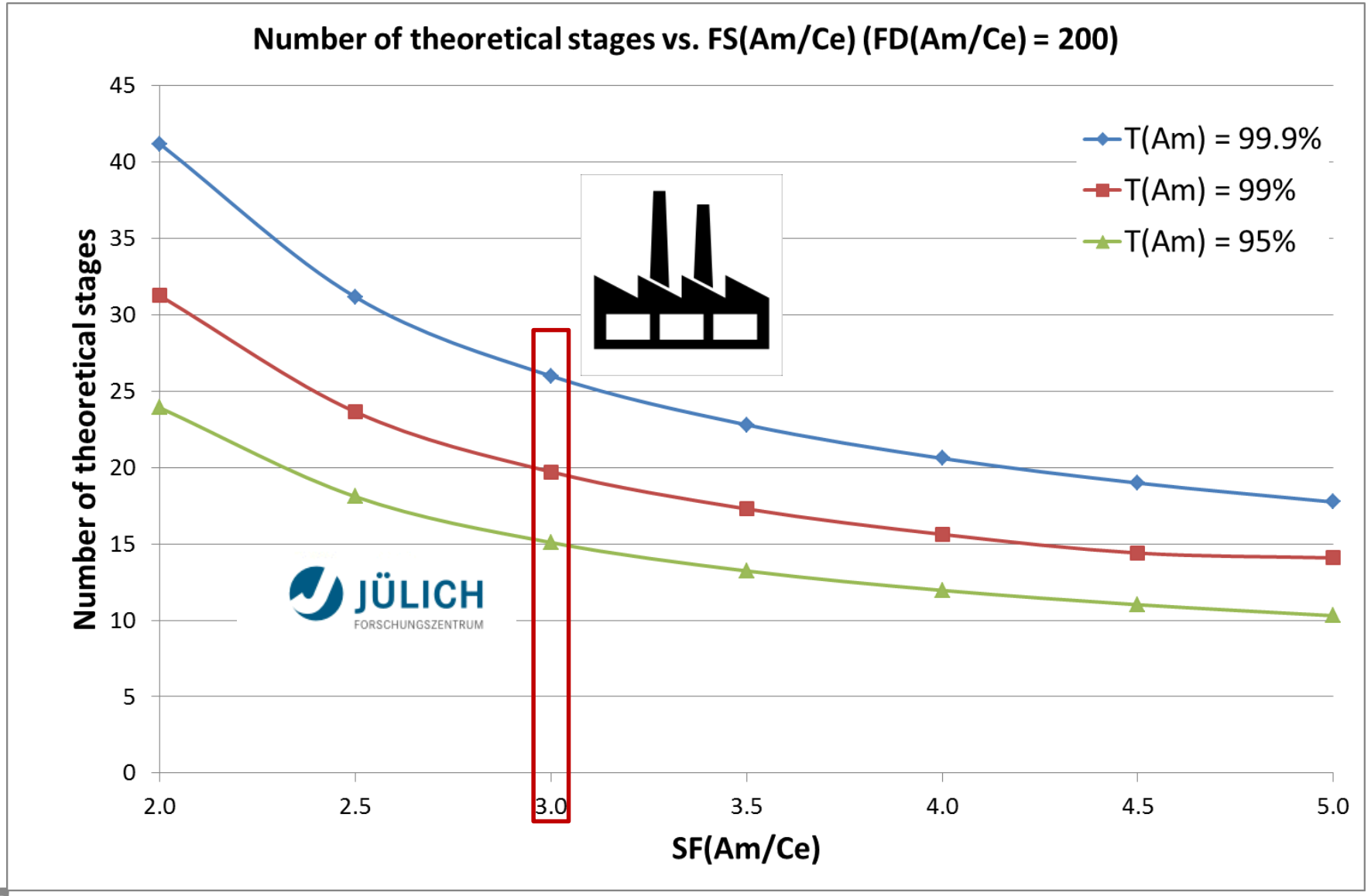
- Am recovery rate : > 90%?, > 95%?, > 99%?
- DF(Am/Cm) > 500 ?
- Ln / (Am + Ln) < 5% ?
- Number of stages

Number of stages
hot test ≠ industrial process

Which DF(Am/Ln) ?



Am recovery vs. number of theoretical stages and SF(Am/Ce)



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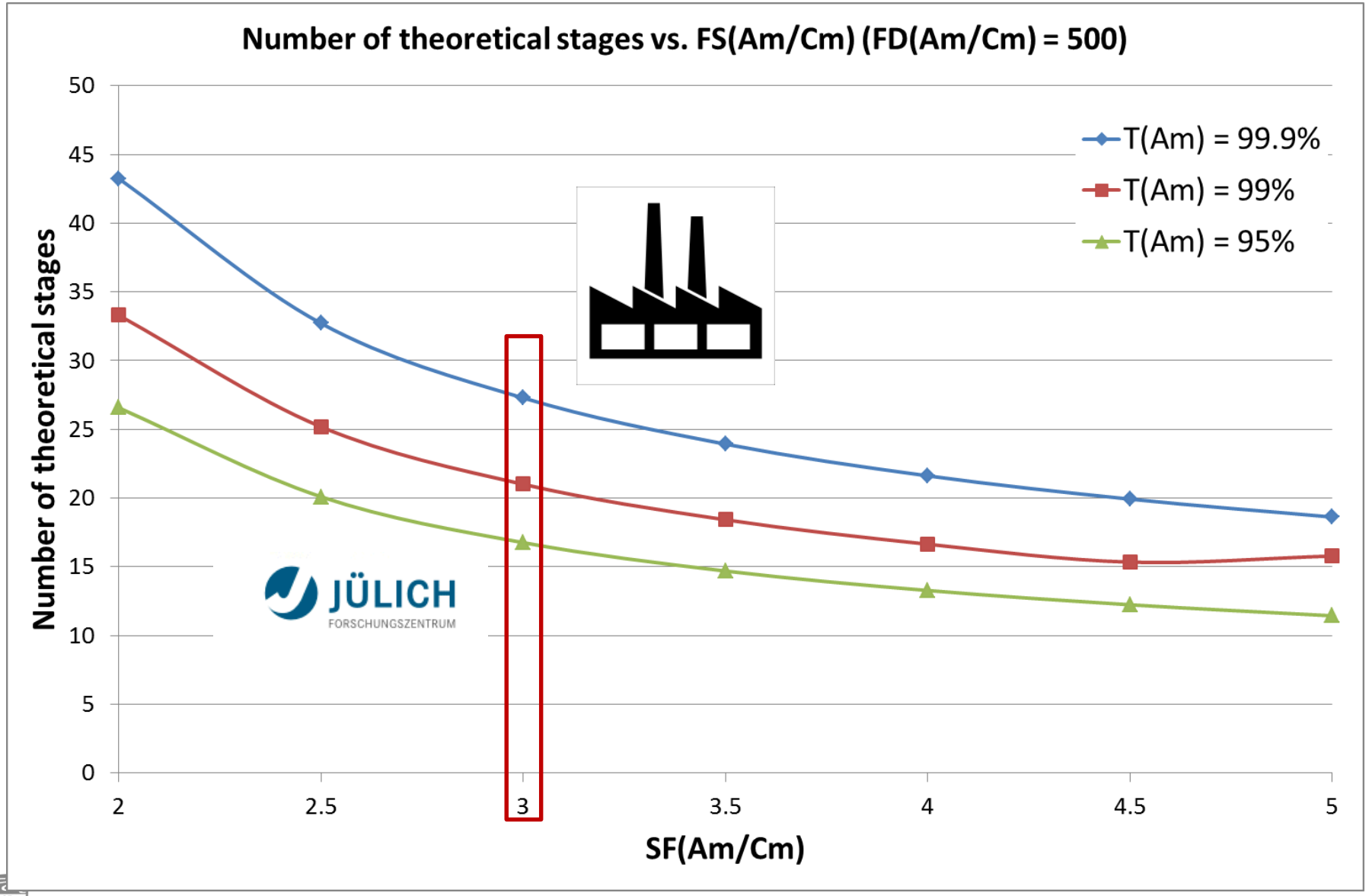
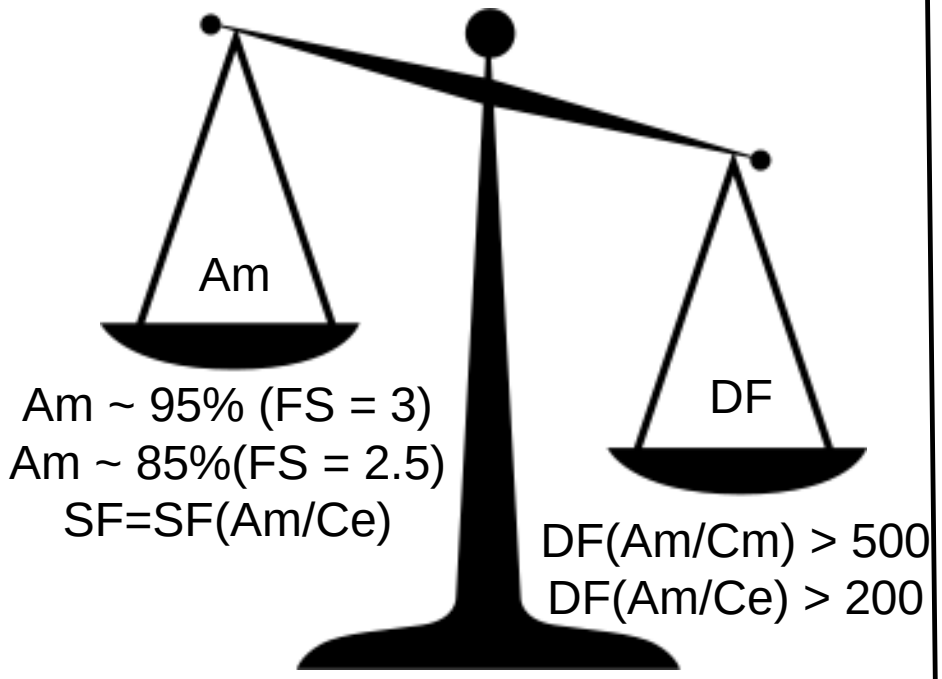


Target performances

JÜLICH FORSCHUNGSZENTRUM

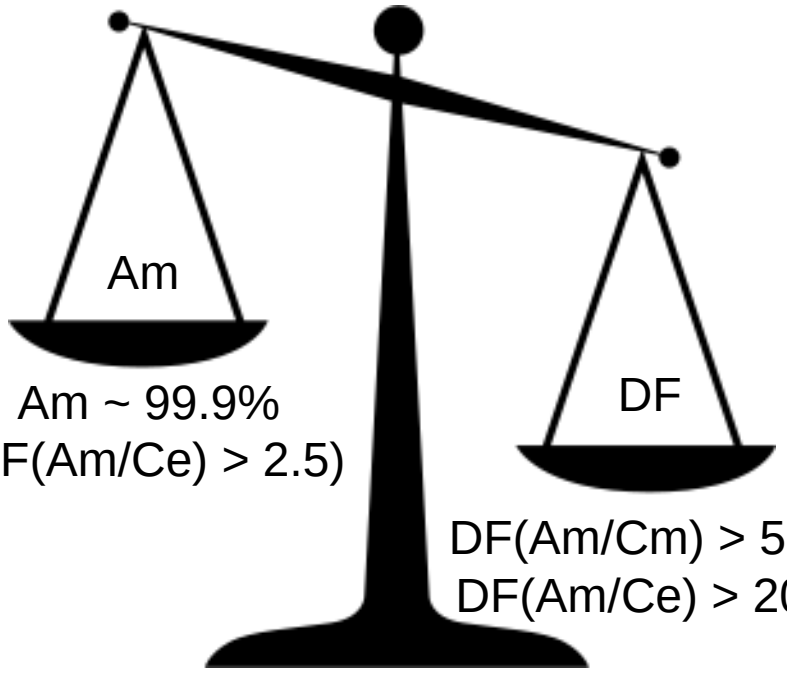
Number of stages = 16



Am ~ 95% (FS = 3)
Am ~ 85%(FS = 2.5)
SF=SF(Am/Ce)

DF(Am/Cm) > 500
DF(Am/Ce) > 200

Number of stages = 32



Am ~ 99.9%
(SF(Am/Ce) > 2.5)

DF(Am/Cm) > 500
DF(Am/Ce) > 200

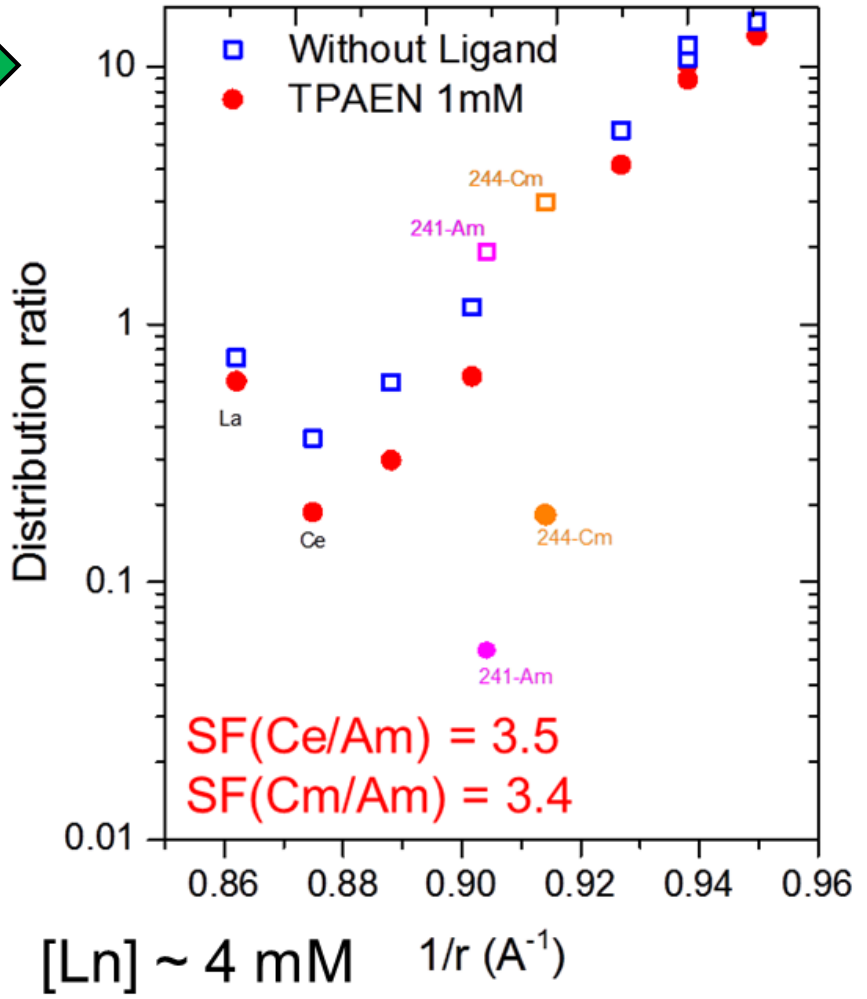
Modelling implemented in the PAREX code

- Experiments used (2+2)
- Assumption : diluted solutions (no salt, no high nitric acid concentration) => not necessary to take into account the deviation from ideality (chemical activities)
- Phenomenological model

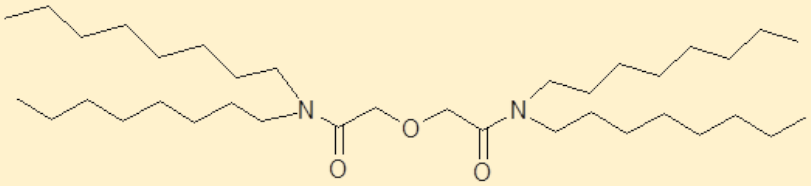
$$M^{3+} + 3NO_3^- + E \Leftrightarrow \overline{M(NO_3)_3E}$$

$$\left[\overline{M(NO_3)_3E}\right] = K[M^{3+}][NO_3^-]^3[\bar{E}]$$

- First model = a lot of improvements to be made but a good way to determine a list of further experiments



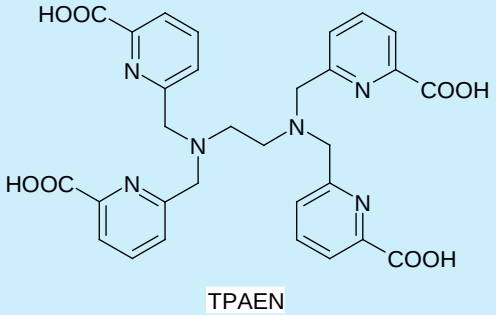
Modelling implemented in the PAREX code



$M(NO_3)_3TODGA$

MH_4TPAEN (1:1)

H_6TPAEN
H_4TPAEN
H_3TPAEN^-
H_2TPAEN^{-2}
$HTPAEN^{-3}$
$TPAEN^{-4}$

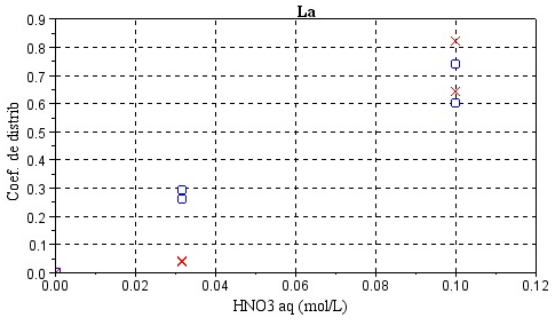


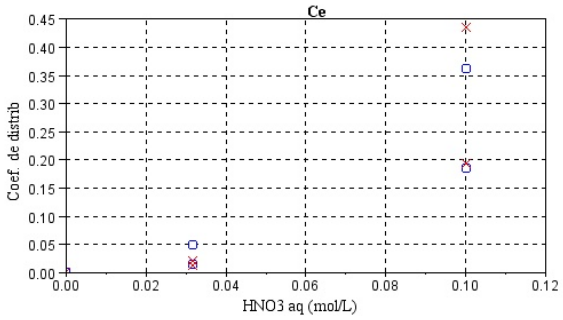
TPAEN

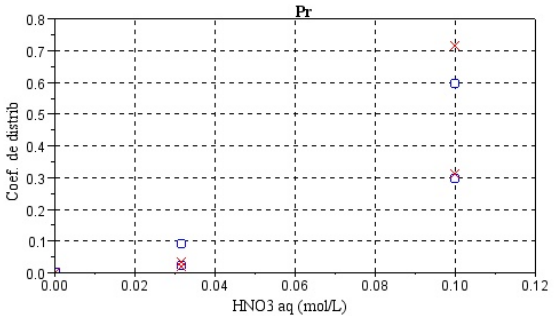
Modelling implemented in the PAREX code

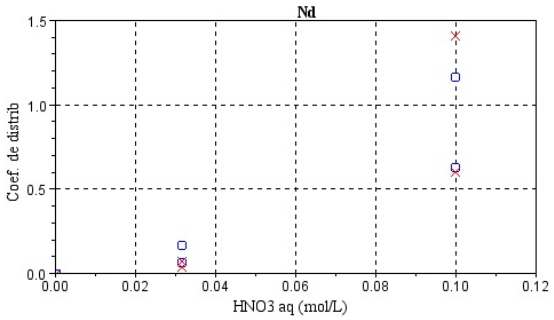
Log(K)	H_n	Am	Cm	La	Ce	Pr	Nd	Sm	Eu	Gd
M(NO3)3TODGA (modelling) (2 experiments)	n=1: -0.27 n=2: -1.8 n=3: -3.82	4.0	4.2	3.6	3.3	3.5	3.8	4.5	4.8	4.9
M(H4TPAEN) (modelling) (2 experiments)	n=6: 2.8 n=4: 3.2 n=3: 3.9 n=2: 5.1 n=1: 7.8	6.9	6.6	4.8	5.4	5.4	5.4	5.1	4.9	4.7
M(H4TPAEN) (experimental)	n=6: 2.8 n=4: 3.2 n=3: 3.9 n=2: 5.1 n=1: 7.8	6.1	4.3	2.79	2.57	3.19	3.26	2.97	2.75	-

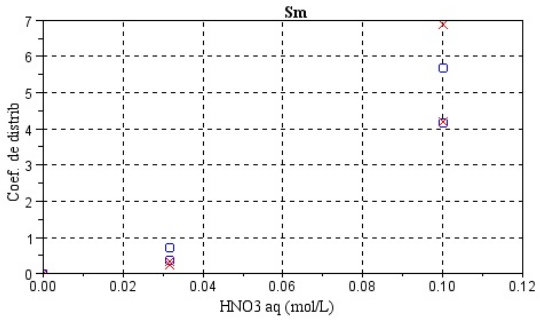
Modelling implemented in the PAREX code

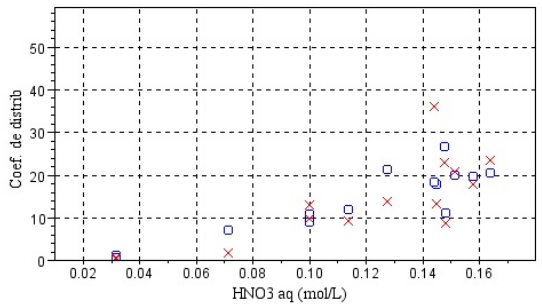


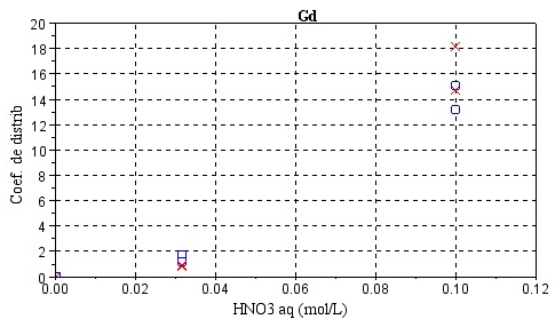


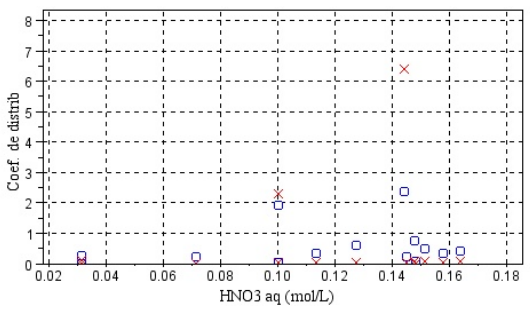


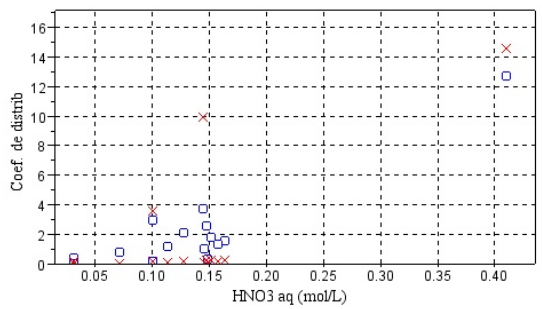




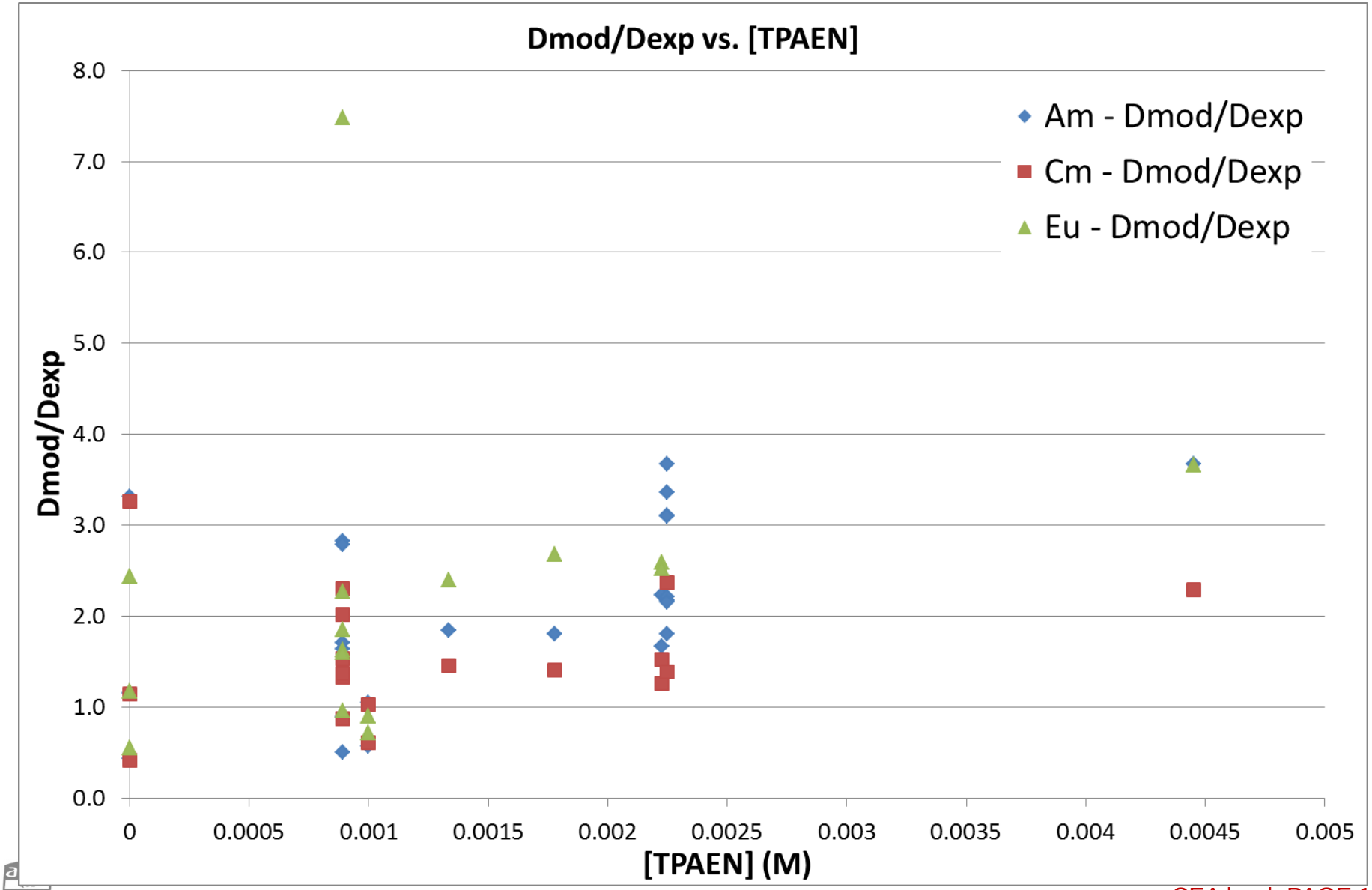






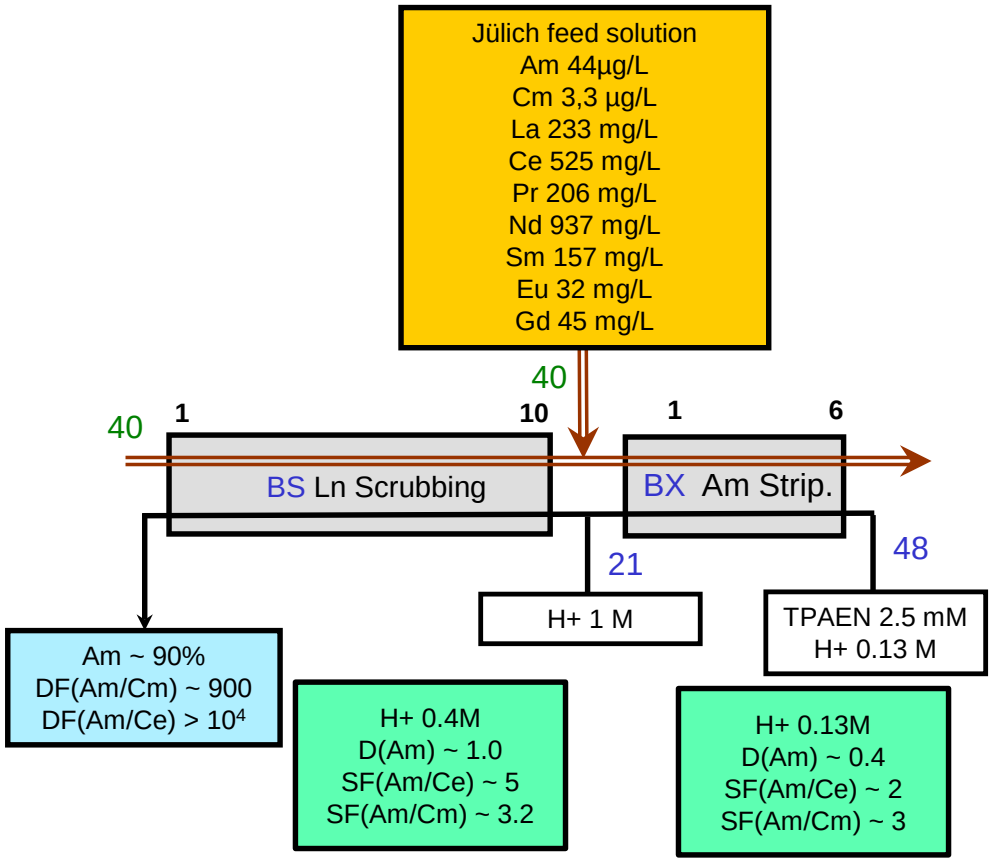


Modelling implemented in the PAREX code

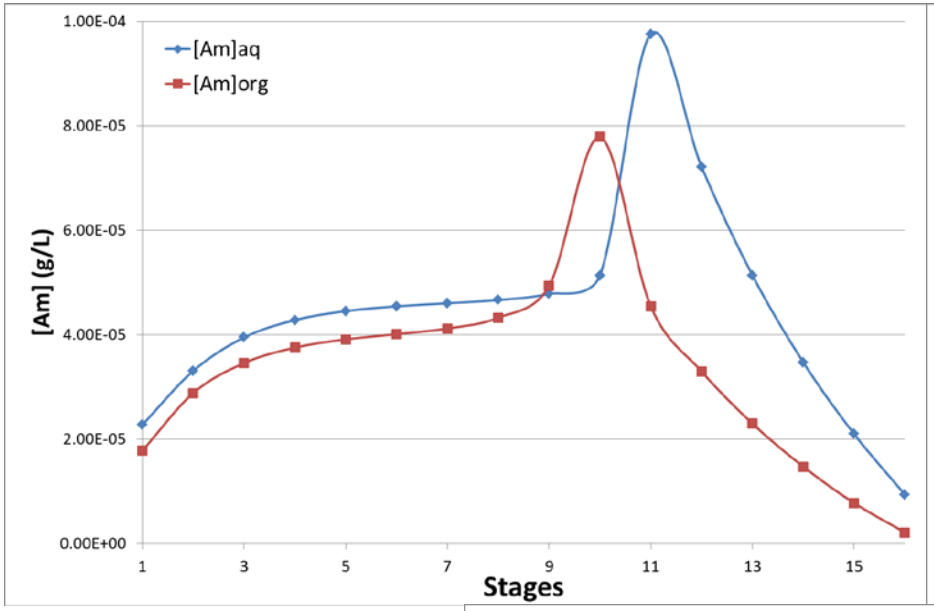


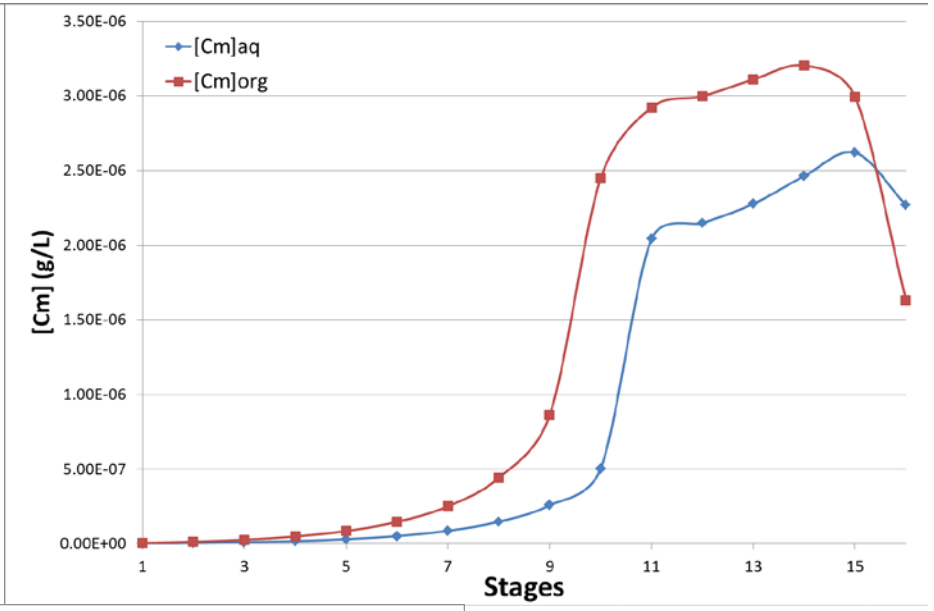
Suggestion of a flowsheet

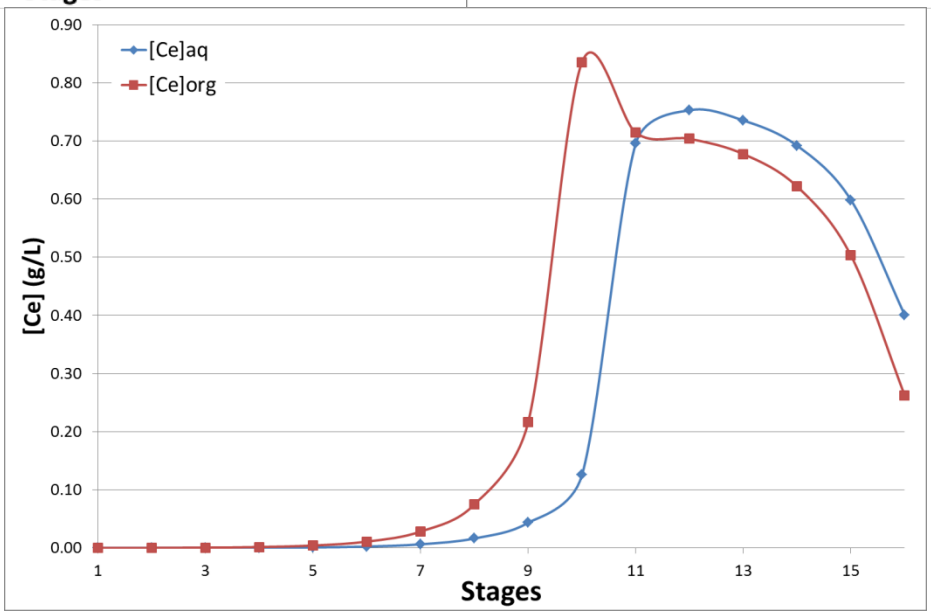
- Use D(M)= f([H+]): BX lower D / BS higher D



Suggestion of a flowsheet







Conclusions

- Difficulties to simulate the last experimental data with macroconcentrations of Ln.
- A first flowsheet made, but necessary to improve this model with new experimental data :
  - Data at equilibrium : D(M), [M]total, ([M]aq, [M]org)
  - 2 families of Ln : La -> Nd and Sm -> Gd. Experiments with one representative from each family (Ce and Eu?) and then with all elements (Am, Cm, Ln)
  - [Ln] : 1-5 mM (BS) to 25 mM (BX)
  - [H+] : 0,1M (BX) to 0.5 M (BS)
  - TPAEN : detemine the solubility limit (2.5 mM?)
  - [TPAEN] : 0mM (extraction by TODGA alone) to solubility limit of TPAEN

TPAEN = 0 mM	[Ln] = 1 mM	[Ln] = 5 mM	[Ln] = 15 mM	[Ln] = 20mM	[Ln] = 25 mM
[H+] = 0.1 M					
[H+] = 0.15 M					
…					
0.5 M					

Conclusions

- Implementation of kinetics aspects and temperature
- Suggestion of a consolidated flowsheet for Jülich hot test
- Sensitivity studies

- Study to reduce concentrations of La and Ce in loaded solvent (optimization with Am recovery rate) ?

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