Downstream effects of aqueous effluents generated by the EXAm separation process

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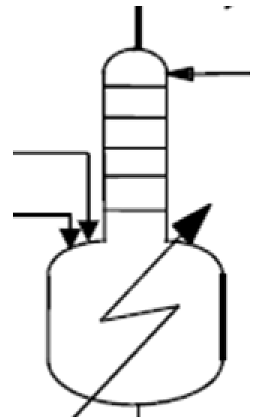
Objectives

- determine whether the treatment of EXAm reagents meets output specifications

Aqueous phases from the EXAm process (acidic media)

Aqueous phases from the solvent treatment (acidic media)

Aqueous phases from the AmO_2 production (acidic media)

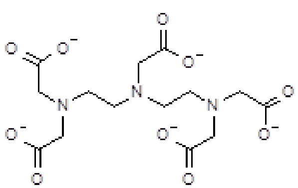


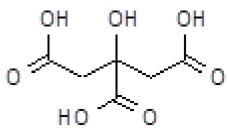
Tank : Radiolysis /hydrolysis

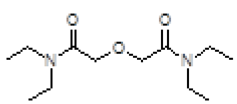
Vitrification
TOC < 550 mg/L

Nitric acid recycling
No specification (TOC)

Tritiated water
COD < 1 g/L
(60 tons a year)

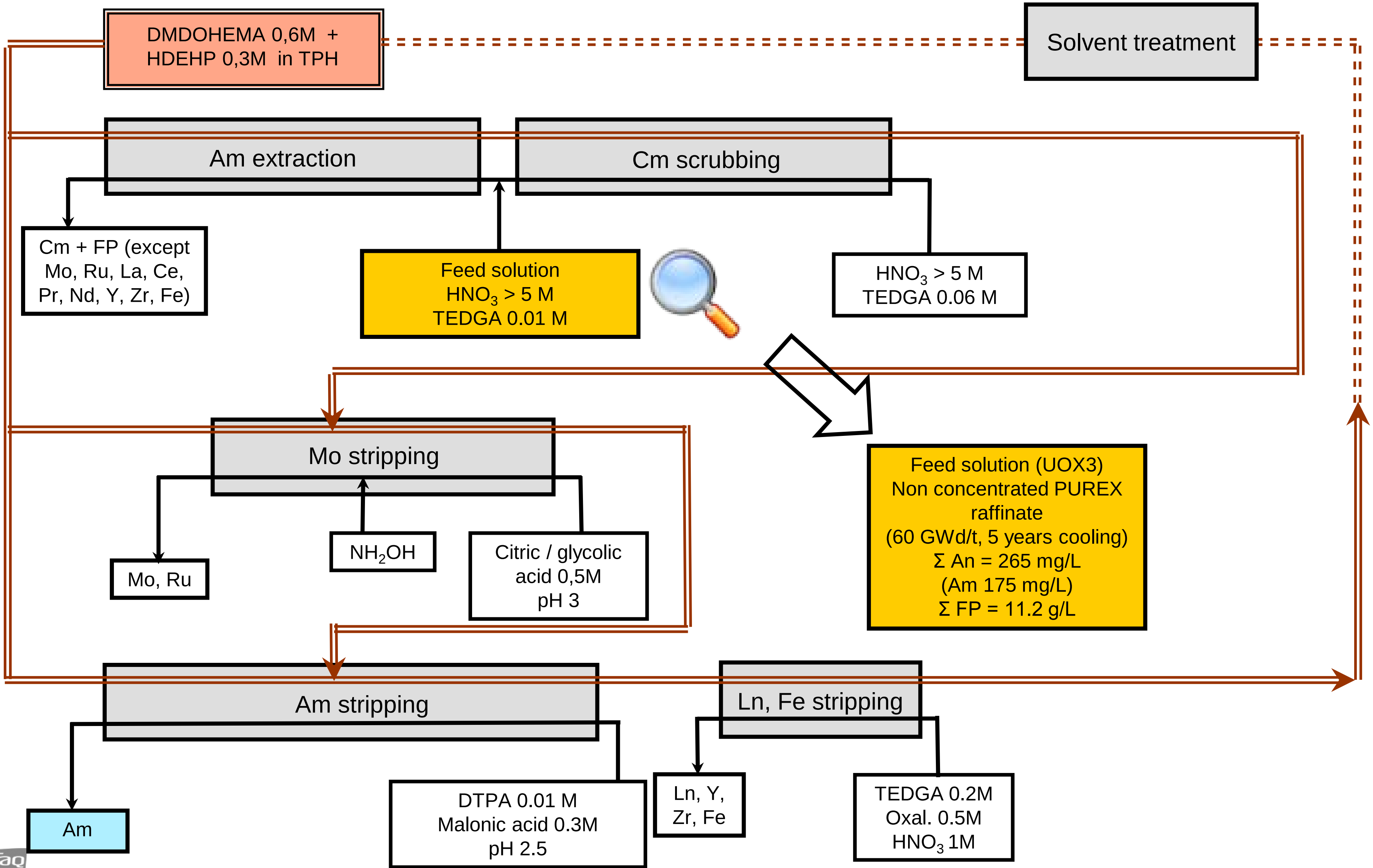




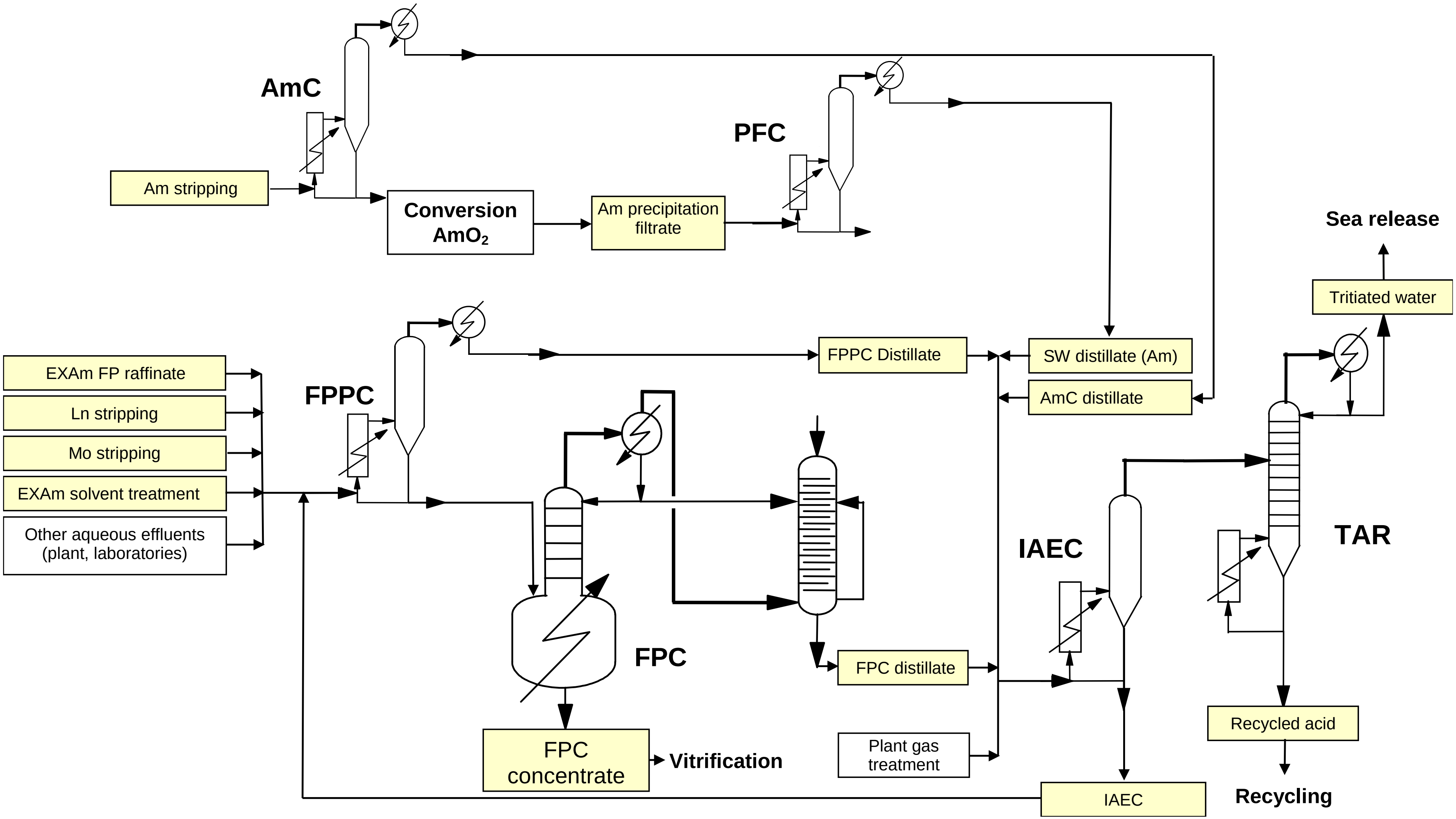


NH_2OH

EXAm flowsheet studied



Aqueous effluents management flowsheet

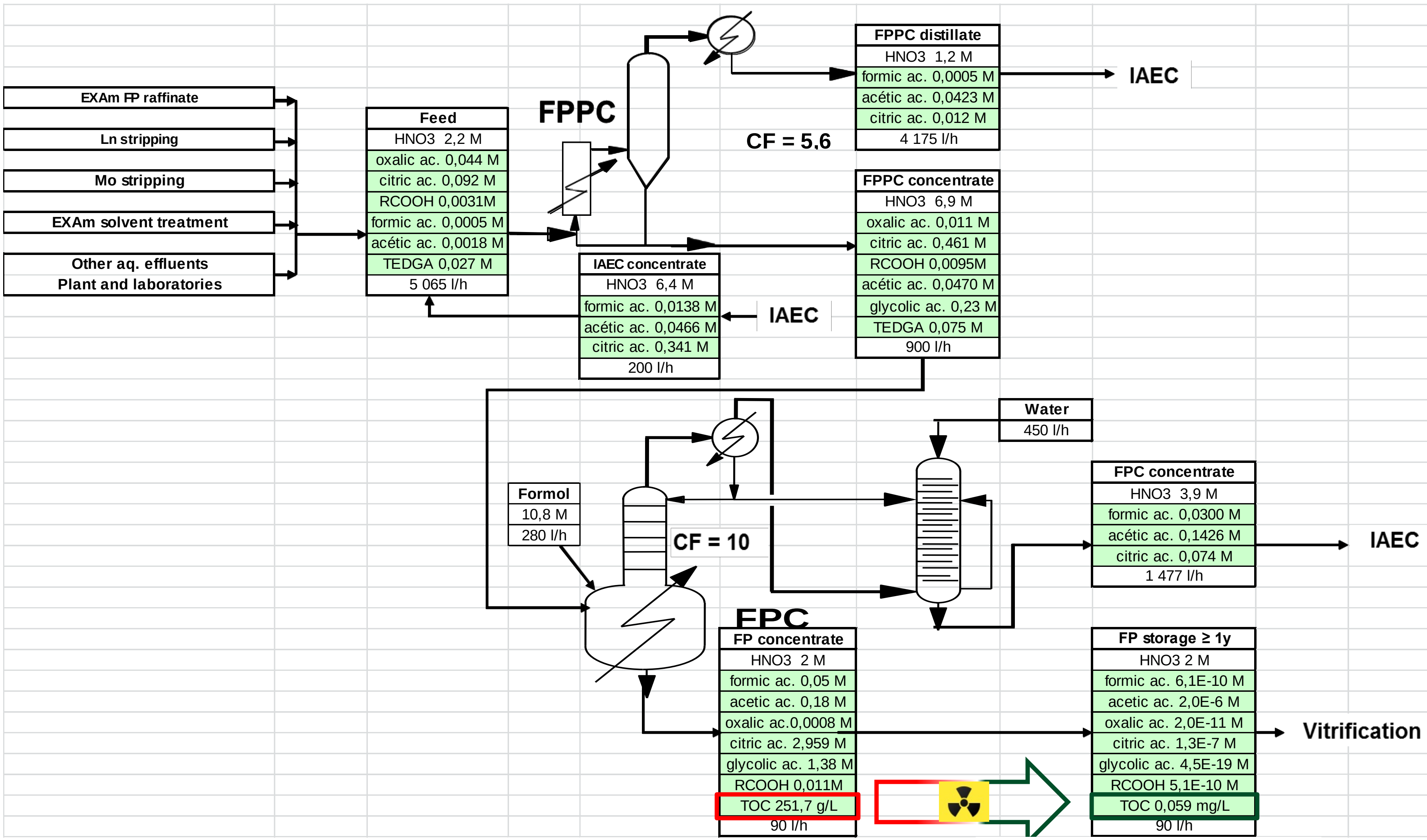


Simulation methodology

- Distribution to different flows : α (distillate/ concentrate separation distribution)
- Destruction :
  - TOC = Σ C atoms
  - Chemical molecule -> intermediate molecules -> CO_2 + degradation product (fewer or no C atoms) (thermolysis, denitration, radiolysis)
  - k = destruction kinetics
  - Grouped destruction sometimes > elementary destruction
  - FP flow : elementary destruction (may be pessismistic)
  - Am production concentration + co-precipitation + filtrate concentration : elementary and grouped destruction
  - Not studied : TEDGA (TODGA data used) + carboxilic acid (propionic acid, butyric acid, Fornet data used)
  - Example of malonic acid :


Phenomena	k	Degradation products
Thermolysis	0	-
Thermolysis + denitration	7.5 h^{-1}	Formic / acetic / oxalic / CO_2
Radiolysis	TOC = 0.00036 kGy^{-1} 0.1M HNO_3	Degradation products + CO_2

FP PreConcentration (FPPC) + FP concentration (FPC)



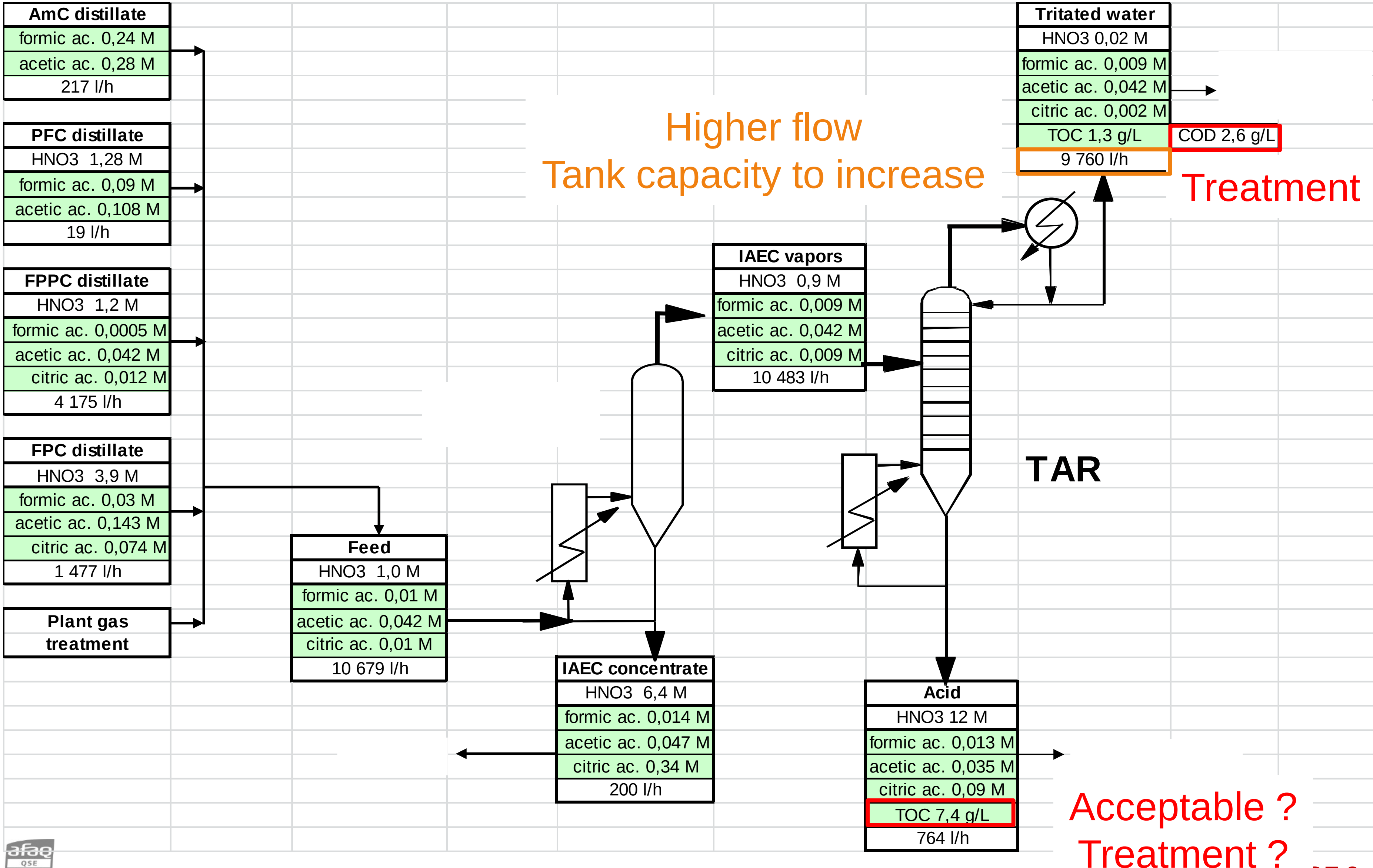
FP precipitate ?

TEDGA ≈ 33% TOC

TOC < 550 mg/L

(4.5 months)

IA Effluents Concentration (IAEC) + Tritiated Acid recycling (TAR)



Conclusions (non concentrated PUREX raffinate)

- Nothing prohibitive but :
  - $[TOC]_{vitrification}$ < 550 mg/L due to radiolysis.
  - $[TOC]_{recycled\ acid}$ higher than current values (no specificiation). A treatment may become necessary in case of TOC limitations.
  - $[TOC]_{sea\ release}$ higher than current regulatory requirements. A treatment should be studied. Higher capacity tank will be necessary.
- Assumptions to be refined : effect of grouped destructions ? Destruction of TEDGA? Destruction of carboxilic acid?

EXAm on a concentrated PUREX raffinate (CF = 3.5, [FP] ~ 36 g/L)

- Main changes :
  - HDEHP from 0.3M to 0.45 M.
  - HEDTA added (complexing reagent for Pd)
  - Flow rates adapted to feed solution

- 2 flowsheets simulated (with and without TEDGA) with same target performances (2 models but modelling assumptions are not detailed here)
  - TEDGA : Am extraction – Cm scrubbing (40%) + Ln stripping (60%).
  - Models for flowsheet without TEDGA less terminated
  - Same feed solution flow rate (50 mL/h)
  - Higher number of stages compared to flowsheet with TEDGA (162 stages instead of 72)
  - Higher organic and aqueous flow rates compared to flowsheet with TEDGA

EXAm on a concentrated PUREX raffinate
(CF = 3.5, [FP] ~ 36 g/L)

Flow (M/h)	EXAm (with TEDGA)	EXAmCO (with TEDGA)	EXAmCO (without TEDGA)
TEDGA	134.5	152.3	0.0
HEDTA	0 (*)	28.2	48.8
DTPA	2.23	10.0	31.2
NaOH	464.0	804.4	1163.0
HNO3	8723	4761	10966
Citric acid	464.0	132.3	249.9
Malonic acid	66.9	97.0	308.7
Oxalic acid	222.5	235.2	3528

(*) HEDTA not used yet

EXAm on a concentrated PUREX raffinate (CF = 3.5, [FP] ~ 36 g/L)

		EXAm (with TEDGA)	EXAmCO (with TEDGA)	EXAmCO (without TEDGA)
Vitrification (FP)	Flow rate (L/h)	90	90	90
	TOC (mg/L)	0.059	0.063	0.008
Recycled acid (12M)	Flow rate (L/h)	764	516	1209
	TOC (mg/L)	7.4	5	4.5
Sea release (tritiated water)	Flow rate (L/h)	9760	4337	13488
	COD (g/L)	2.6	7.0	2.9
	Annual COD (t/year)	121.8	145.7	187.8

Trends on the impact of a concentrated PUREX raffinate Am separation on aq. Effluents management

- Vitrification (TOC)
  - Meet specification in every case (lower without TEDGA).
  - No significant impact of concentration.

- Acid recycling (TOC)
  - Same order of magnitude, lower with EXACO. A treatment may become necessary in case of TOC limitations (same conclusion as a non concentrated PUREX raffinate).
  - BUT flow rates are significantly different. Removing TEDGA lose the benefit to concentrate feed solution.

- Sea release (COD)
  - Treatment remains a challenge in every case. EXACO with TEDGA closest to annual COD but more concentrated
  - COD(EXACO with TEDGA) > COD(EXACO without TEDGA) ~ COD(EXAm). Removing TEDGA lose the benefit to concentrate feed solution.

Removing TEDGA lose the benefit to concentrate feed solution for acid recycling ans sea release. Necessary to find another complexing reagent for Ln stripping.
Meeting COD specifications will be the main challenge for aqueous effluents management

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