R&D Opportunities in Head-end Processing and Off-gas Management

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Outline

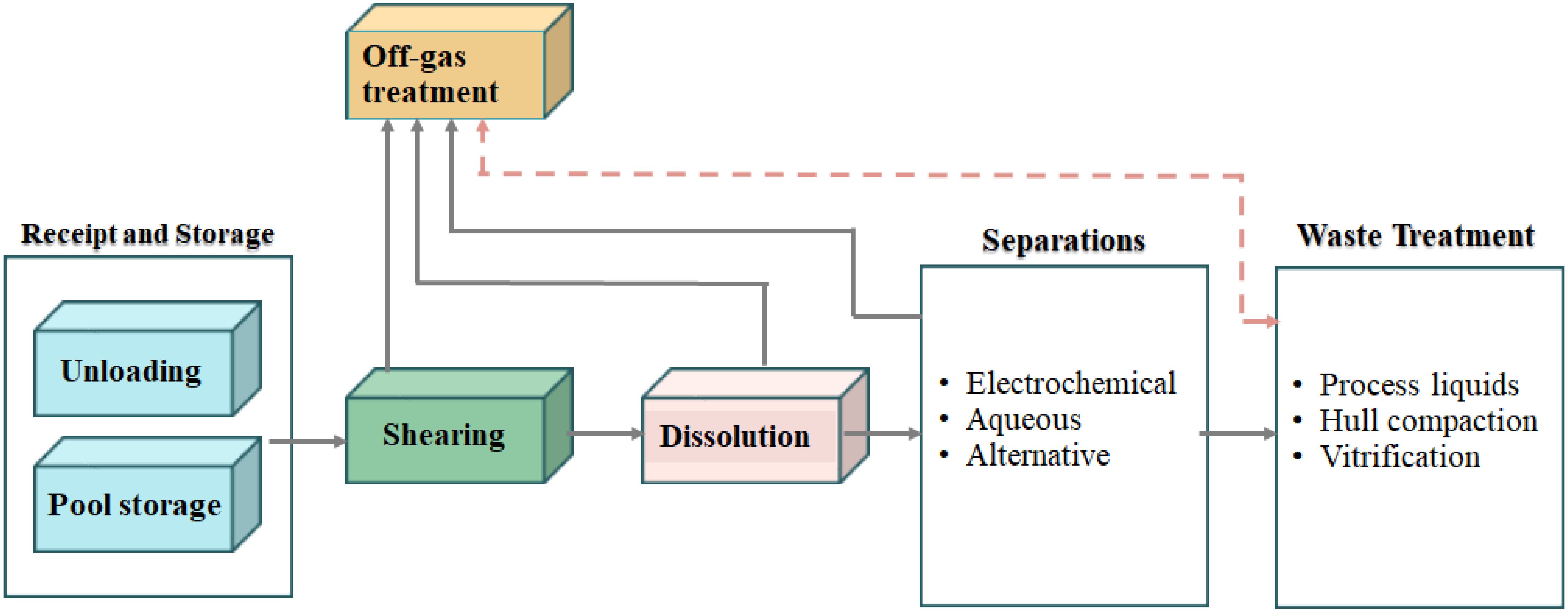
- What is head-end processing and off-gas management?
- Why are we interested in these aspects of fuel processing?
- How do these processes work within a facility?
- Where are there opportunities for improvement?

Head-end: handling of fuel assemblies prior to beginning chemical separations.

- Pool storage
- Disassembly or hardware removal
- Shearing/Fuel meat exposure

Off-gas: All gaseous process streams within a facility requiring treatment.

- Contains volatile fission products, including noble gases
- Radioactive emissions must be managed to comply with regulatory discharge limits



Access to cost information can be challenging, but we do know the following:

- **Shielded hot cells** are required for handing spent fuel until activity is reduced through chemical separations
- **Fuel assemblies for Gen III+ reactors are long**
  - *Westinghouse 17X17 ≥ 12 feet active fuel length*
  - *PWR assembly length > BWR assembly length*
- The head-end portion of the plant is typically **less flexible**, relies on mechanical operations, and can be a limiter in terms of **operational availability**
- Requires **remote maintenance**

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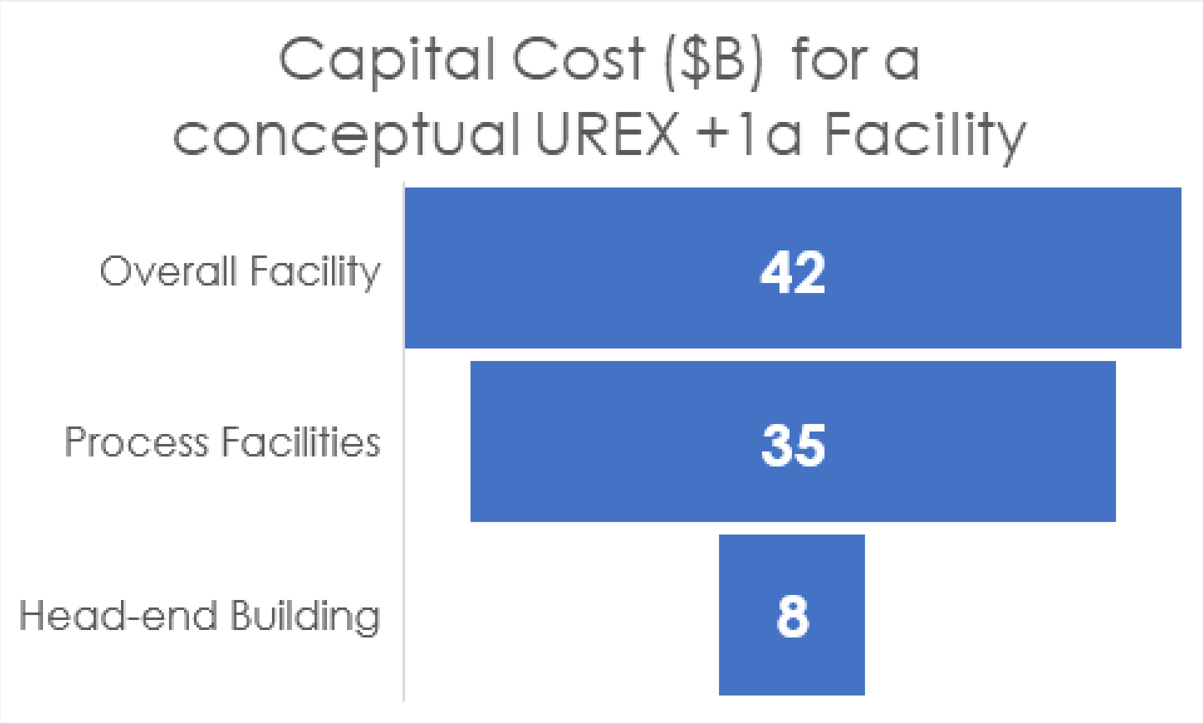
Consider a conceptual UREX +1A plant:

Plant specifications:
- UREX +1a separations design
- 3000 MT/y throughput

Head-end building contained:
- Fuel receipt
- Fuel storage (SFP)
- Fuel shear
- Fuel dissolution
- Dissolver off-gas treatment
- Hulls rinse/compaction
- Tc management



Dixon, et al. Advanced Fuel Cycle Cost Basis – 2017 Edition. NTRD-FCO-2017-000265. September 2017.

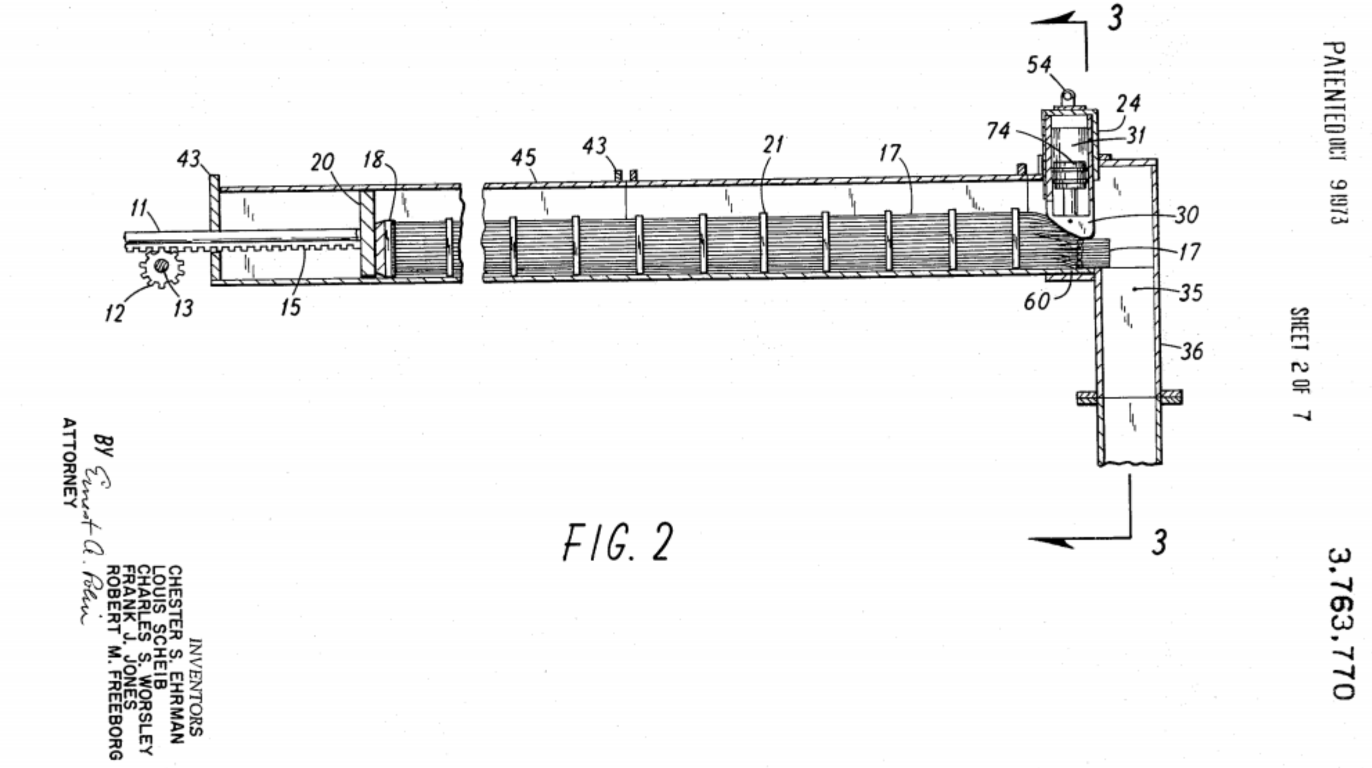
Things that decrease capital cost can also reduce operating and maintenance costs.

- Decrease capital through:
  - Decrease of overall facility footprint, but especially shielded space
  - Improved equipment design and fabrication
- Decrease shielded footprint:
  - Early volume reduction
  - Early decontamination to glovebox-acceptable activity levels
  - Choose equipment with small footprints, choose flowsheets with fewer, smaller, and less shielded operations
- Large physical footprints *for aqueous*:
  - Mechanical shear
  - Dissolver off-gas treatment
  - FP/HLW waste treatment and associated off-gas

What's the baseline for head-end processing?

- **Fuel receipt**
- **Fuel storage** – available wet storage can vary widely across designs and is important in costing the head-end
- **Shearing of UO_x fuel**
  - Many types of shear designs
    - Bundle shear, single pin shear
  - Alternative blade designs
  - Shearing disadvantages:
    - Creates dust
    - Releases fission products
    - Causes segment crimping

Gullwings
Guillotines
Dogbones?

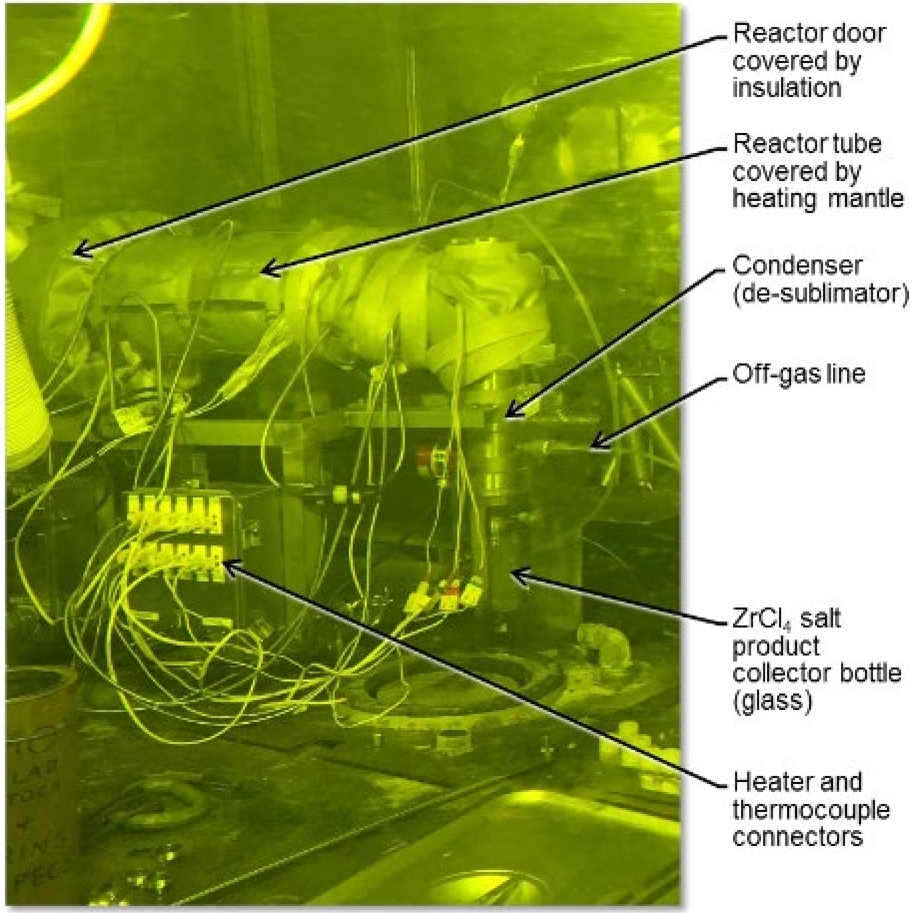


Worsley, C., Jones, F., Ehrman, C., Scheib, L., & Freeborg, R. (1973). *U.S. Patent No. 3,763,770*. Washington, DC: U.S. Patent and Trademark Office.

Alternative to Shearing: Chemical decladding

- Chlorination, hydrochlorination
- Electrolytic removal of zirconium
- Sulfur chlorination digestion
- Dissolution in NH_4F (ZIRFLEX)

➢ Product should eventually be ZrO_2 if disposal is desired. The nuclear grade zirconium is valuable if highly pure and if end-users will tolerate Zr-93 ($t_{1/2} = 1.56 \times 10^6$ y).



Chlorination of 0.5 kg of irradiated hulls in ORNL Hot Fuel Examination Facility

Collins, E. D., Hylton, T. D., DelCul, G. D., Spencer, B. B., Hunt, R. D., & Johnson, J. A. (2018). *Completion of a Chlorination Test Using 250 grams of High-Burnup Used Fuel Cladding from a North Anna Pressurized Water Reactor* (No. ORNL/LTR-2017/327). Oak Ridge National Lab.(ORNL), Oak Ridge, TN (United States).

Questions around chemical decladding…

- What does the reaction vessel look like?
- Is disassembly required?
- Is shearing required?
- Is the uranium-bearing product compatible with downstream processing?

- How does this impact the hulls processing baseline?

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Why off-gas treatment?

- Primary volatile radionuclides: ^{3}H, ^{14}C, ^{85}Kr, ^{129}I
- Semi-volatiles: ruthenium, technetium, others
- Speciation and volatile partitioning varies according to processing flowsheet

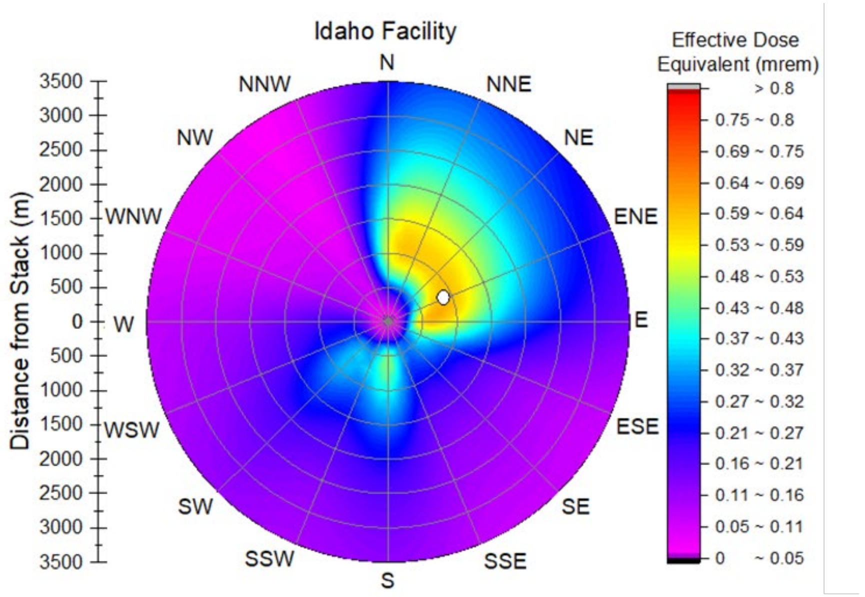
<u>US regulations have two aspects:</u>

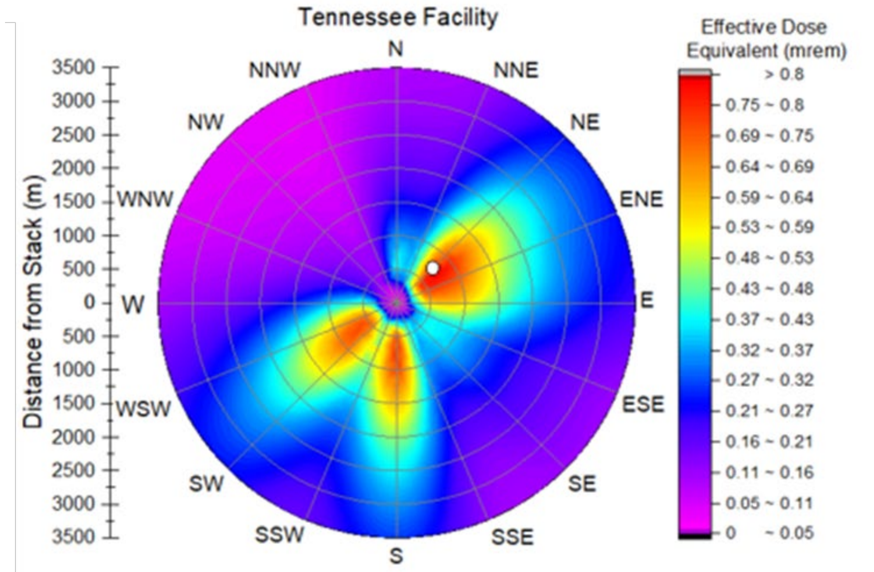
(1) Limit Ci amounts of ^{85}Kr and ^{129}I discharge

(2) Limit dose to public on a whole-body and thyroid basis

➢ Governing regulations are 40 CFR 190, 40 CFR 61, and 10 CFR 20. (EPA and NRC)

S. BRUFFEY et al., *"Requirements and Conceptual Design of Off-gas Systems for the Reprocessing of Metallic Fuels,"* ORNL/TM-2020/1668, Oak Ridge National Laboratory, Oak Ridge, TN (2020).





Air dispersion modeling to determine the dose from iodine emissions

Specific Metrics for Off-gas Systems

- **Iodine-129 release limited to 0.005 Ci/GW_e·y**
  - Expect to retain, <u>at minimum</u>, 99.5 to 99.9% within the facility
- **Kr-85 release limited to 50 Ci/GW_e·y**
  - Expect to capture 70-95% within the facility
- **Total dose to the public from all facility emissions:**

25 mrem/whole body; 75 mrem/thyroid

What is the off-gas baseline for aqueous processing?

Key unit operations:

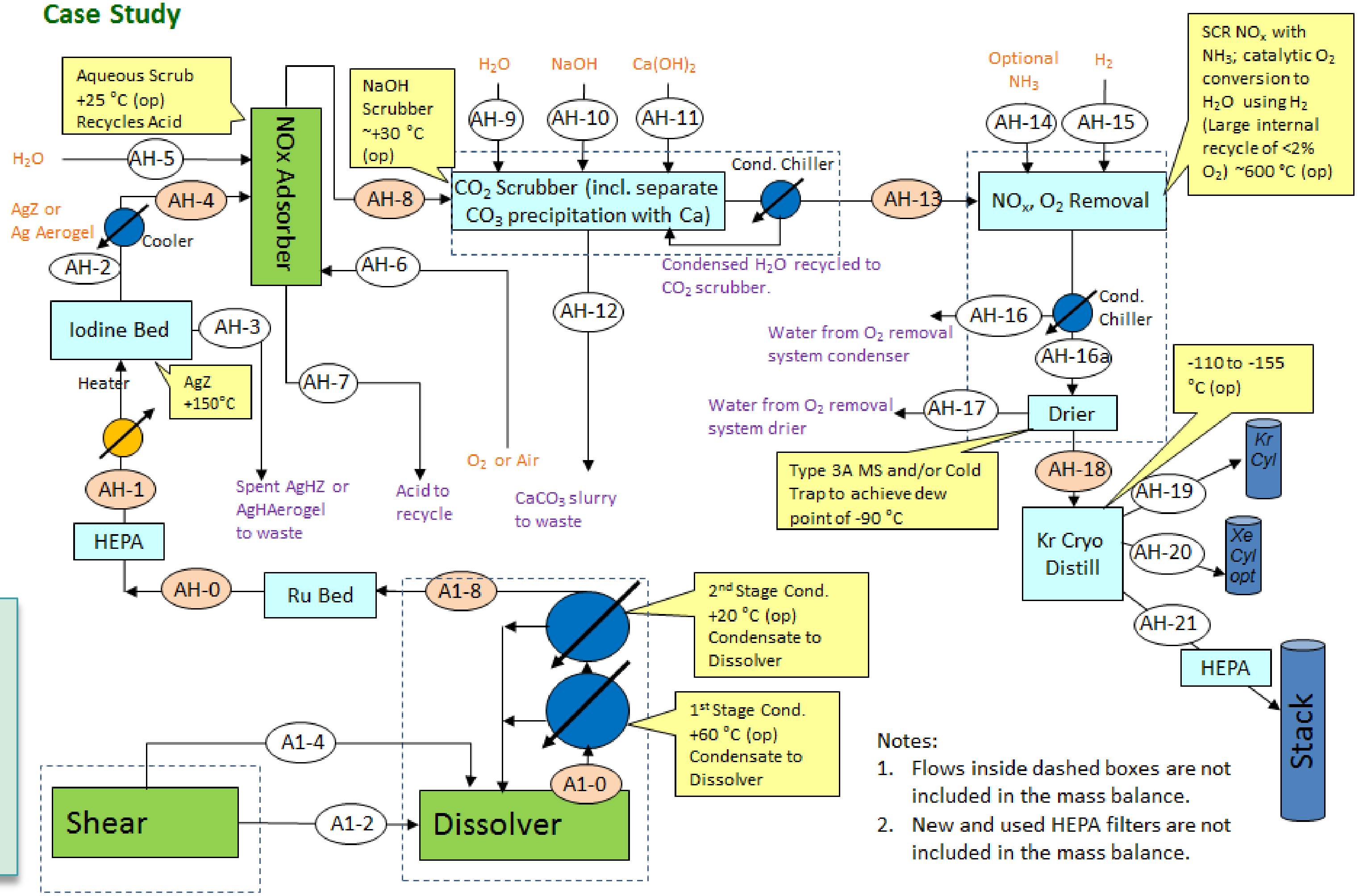
- Ru removal
- Iodine removal
- NO_x removal
- CO_2 removal (optional)
- NO_x, O_2 removal
- Kr/Xe condensation or separations

➢ **Notes:**

(1) Tritium distributes through plant (aq. and gas)

(2) Kr/Xe 1:10 ratio – separations decreases waste by 10×

Aqueous Head-end and Dissolver Off-gas Systems (No TPT) Case Study



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Alternatives have typically included pretreatment:

- **Voloxidation**
(removal of ^3H before dissolution)
- **Advanced voloxidation**
(removal of I and ^3H before dissolution)
- **Direct dissolution**
(advanced volox, no dissolver)
- **High temperature voloxidation**
(OREOX, DUPIC) – removes semi-volatiles

Standard O_2 Oxidation

$UO_2 + O_2 \rightarrow U_3O_8$

Off-gas: ^3H_2O, some semi-volatiles, Kr/Xe
Treatment: Water removal, filtration

Advanced NO_2 Oxidation

$UO_2 + NO_2 \rightarrow U_3O_8 + NO$

Off-gas: ^3H_2O, ^{129}I, Kr/Xe, less semi-volatiles
Treatment: More complex

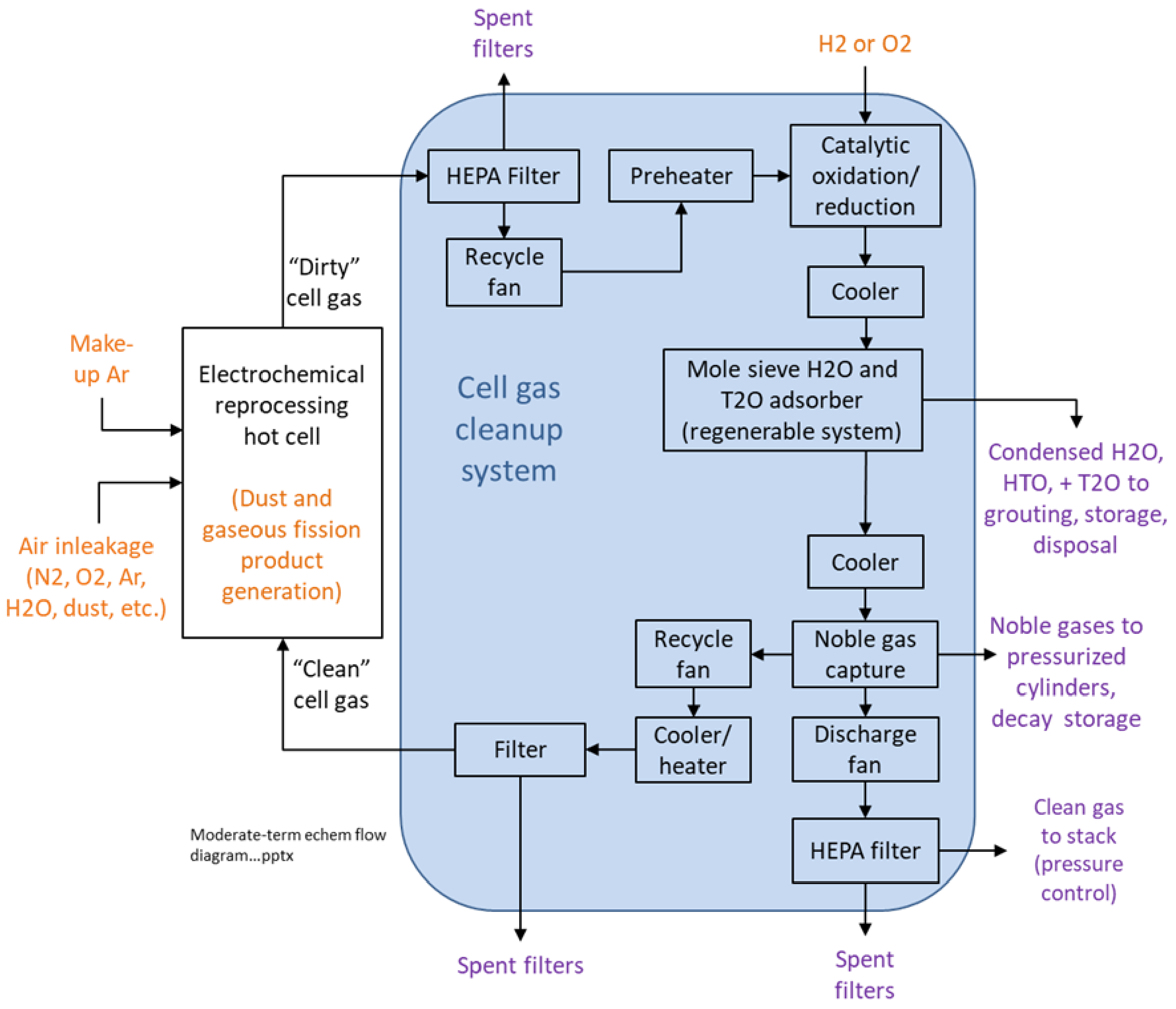
Direct Dissolution

$UO_3 + 5NO_2 \rightarrow UO_2(NO_3)_3NO + 2NO$
$UO_2(NO_3)_3NO$ soluble in organic phase

Off-gas: Eliminate DOG, add Volox-OG

What's the off-gas baseline for pyroprocessing?

- Here, assume the electrorefiner emits to the argon hot cell
- No ^{14}C, less iodine
- Other operations, such as salt distillation, may require additional iodine management



Moderate-term echem flow diagram...pptx

Other off-gas thoughts:

- **Consider other effluents: A balancing act**
  - Control liquid effluents well, higher ceiling on gaseous effluents
  - Control iodine well, limit need for additional controls for other volatiles
  - There is always opportunity for less costly, higher efficiency capture media
- **Consider fuel age:** ^{3}H and ^{85}Kr requirements decrease with fuel age, leaving a higher dose-ceiling for iodine (must always control to Ci/GWd limits)
- **Consider how capture processes can integrate** with waste management
- **Target flexibility:** capture processes that are broadly applicable across multiple processing flowsheets
- **Consider waste forms:** waste volumes generated from off-gas capture are traditionally high volume.
  - Integrate the process with the final waste form
  - Better iodine waste forms are needed, better Kr storage forms are needed

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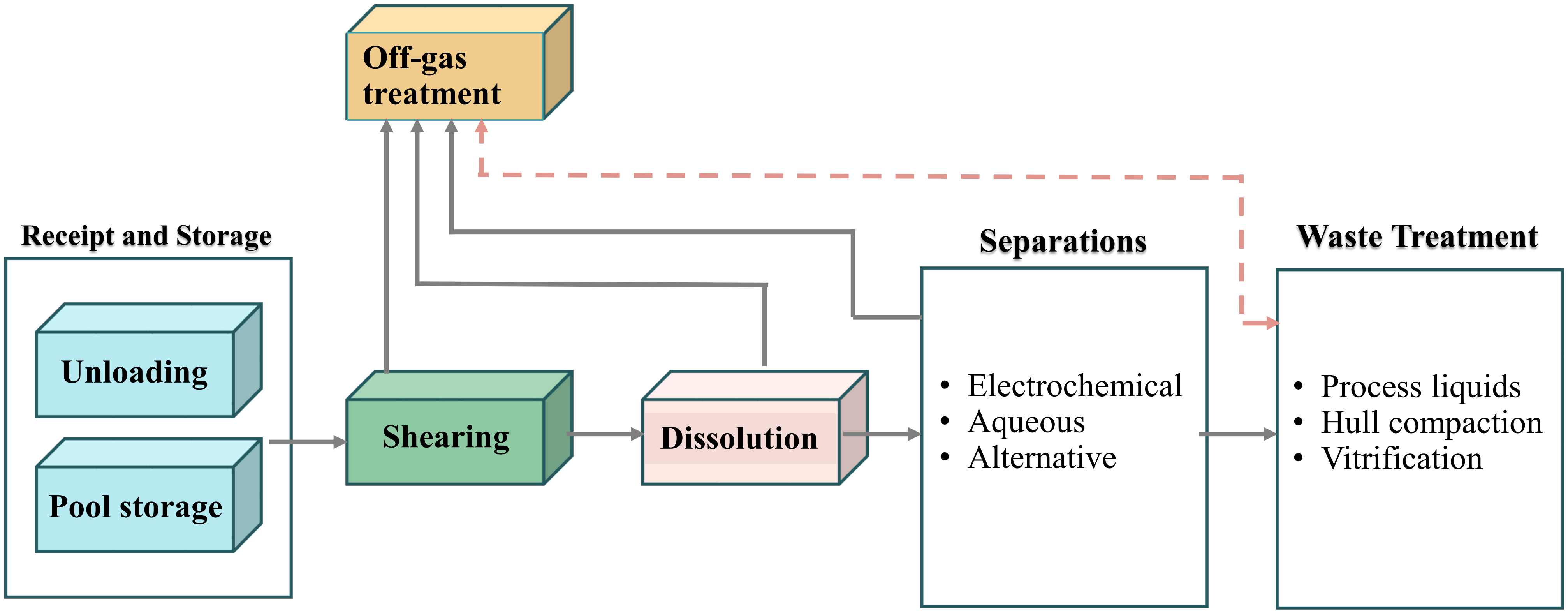
Summary:

- Off-gas/Head-end processing contribute significantly to facility capital cost and O&M
- Alternative head-ends, simplified off-gas treatments, and better secondary waste management can yield improved operations and cost benefit
- Focus on smaller equipment, earlier removal of activity, decreased shielded space

Questions?


Off-gas treatment
Receipt and Storage
Unloading
Pool storage
Shearing
Dissolution
Separations
• Electrochemical
• Aqueous
• Alternative
Waste Treatment
• Process liquids
• Hull compaction
• Vitrification

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