Development of a Novel Dry Chemical Uranium/Molybdenum Separation: Research for a Future Efficient Mo-99 Extraction Process

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On Current Waste Management and the Conversion of HEU to LEU Targets

Waste Management Considerations:[1]

- Conversion of HEU to LEU targets can generate as much as 200% more radioactive waste
- Production of 10,000 6-day Ci/week using LEU targets is expected to produce 15,000 liters per year of intermediate level wastes (ILWs).
    - After cementation, volume can increase to 375,000 liters

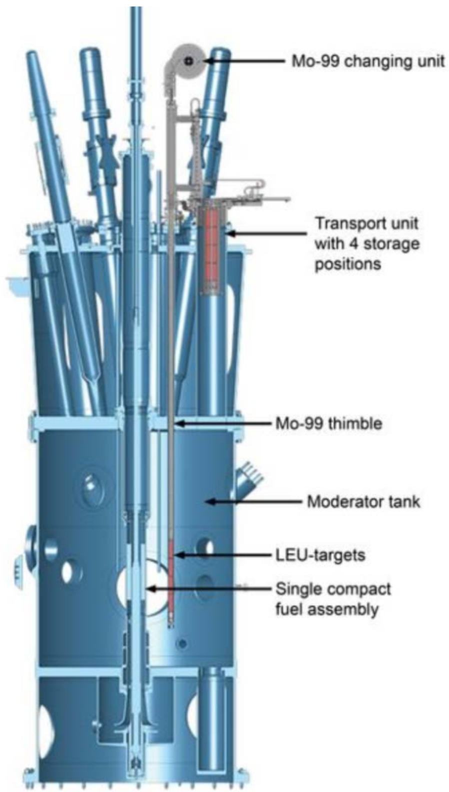
Typical Molybdenum-99 Production Scheme

1. Fabrication of Uranium Targets
    - UAl_x, U_3Si_2-Al, UN, U foil, U metal, UO_2, etc.
2. Irradiation of Targets
3. Dissolution and Moly-99 Recovery
    - Alkaline or Acidic
4. Waste Management
5. Pharmaceutical Delivery

[1] S.K. Lee, G.J. Beyer, J.S. Lee. Nuclear Engineering and Technology. 48 (2016) 613-623.

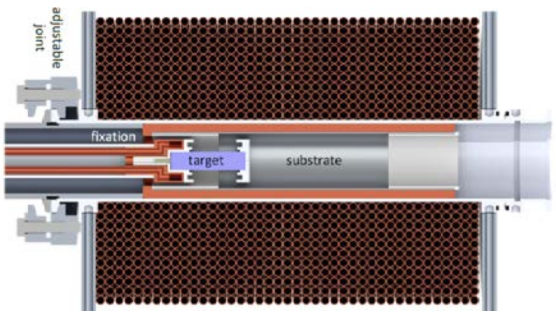
Molybdenum-99 at FRM II

Production

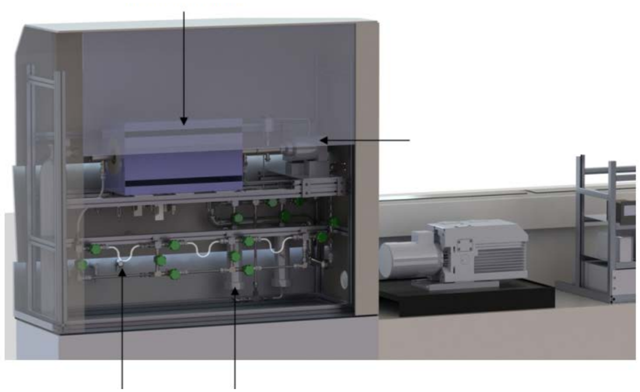


Irradiation Facility

Research

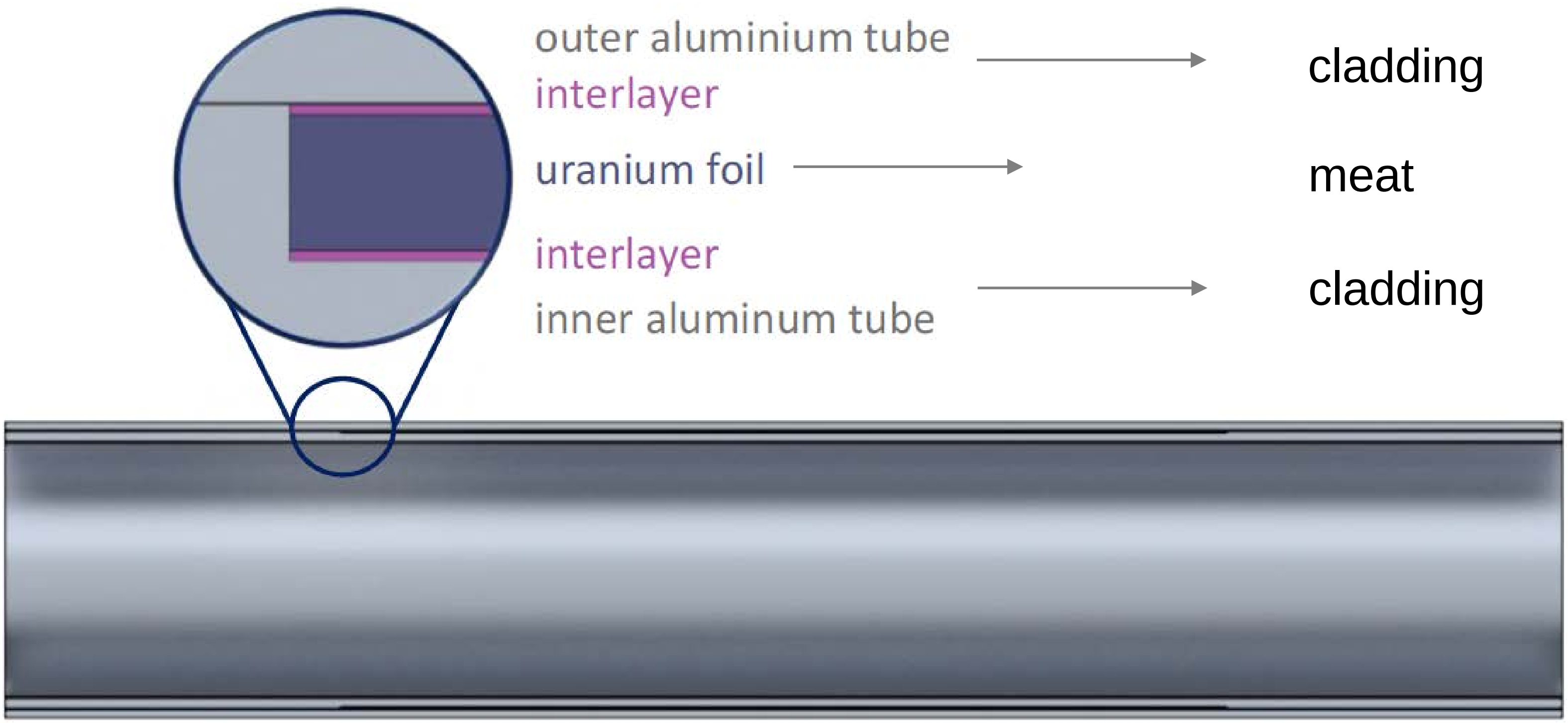


Target Design



Extraction Process

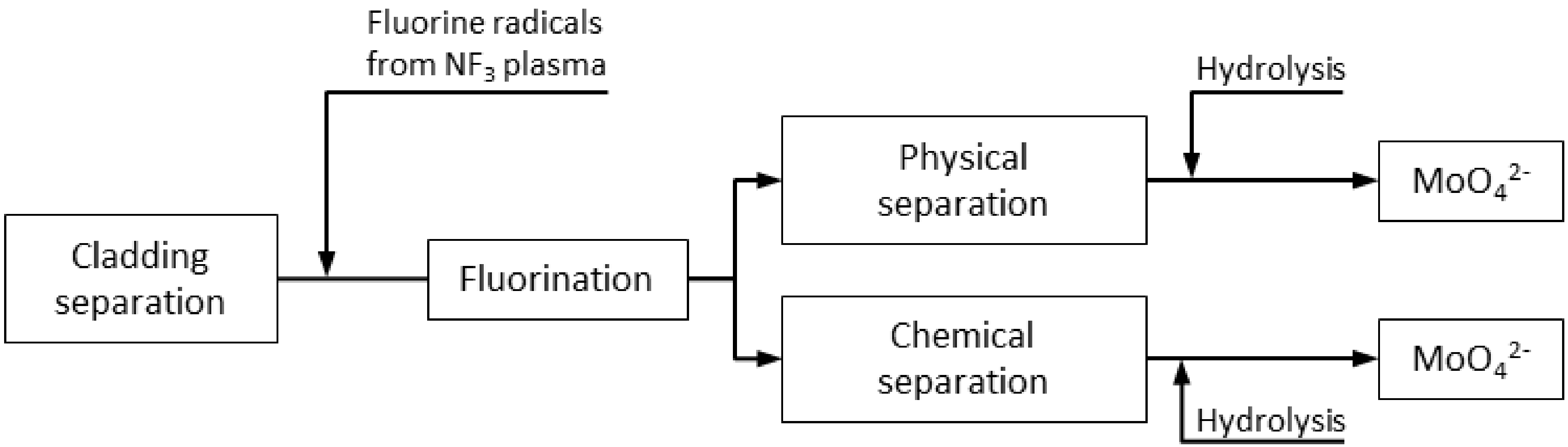
Cylindrical Target Design



With cylindrical design, aluminum cladding can be mechanically separated before dissolution.

- This will lead to decreases in liquid waste production

Proposed Dry Chemical, Plasma-Aided Fluorination Process



- Uranium and fission products fluorinated to higher fluorides with aid of plasma

$$Mo + 6\,F \rightarrow MoF_6$$

- After separation, molybdenum hydrolyzed to MoO_4^{2-} using sodium hydroxide

$$MoF_6\,(g) + 8\,NaOH(aq) \rightarrow Na_2MoO_4\,(aq) + 6\,NaF\,(aq) + 4\,H_2 0$$

The Road to Dry-Chemical Separation

✓ • Cylindrical target production with mechanically separable cladding

✓ • Development and construction of a fluorination line

✓ • Set-up and installation of line system

---→ • Fluorinaiton experiments

• Surrogate material production

• Combination with a plasma vapor deposition process

Production of a Surrogate Material

Element	Mass [mg]	m.p. [°C]	b.p. [°C]
Xe	35,5	-111,7	-108
Zr	28,5	1857	4409
Ce	25,2	798	3426
Mo	17,5	2623	4639
Nd	16,7	1024	3100
Ba	15,5	727	1640
Ru	14,3	2334	4150
Sr	11,8	777	1382
Cs	11,2	28,5	671
La	9,3	920	3457
Pr	6,47	930,8	3512
Y	5,03	1522	2930
Te	4,87	449,5	987,8
Tc	3,61	2204	4265
Pu	3,32	639,4	3230
I	3,25	113,7	184
Kr	3,06	-157,4	-152
Rb	3,04	39,3	688
Sm	2,23	1072	1900
Np	1,57	639	3902
Pm	1,29	1080	3000
Pd	1,10	1555	2960

- Inactive (excluding uranium) surrogate material containing most important elements
- About 10 fission products to be taken into consideration
- Investigate the behavior of surrogate materials in plasma line
- Process surrogate materials through the most promising physical and chemical separation techniques

Chemical Investigations after Fluorination

- Higher fluorides of uranium and molybdenum expected from plasma-induced fluorination.

HF																	He
						MnF_3, MnF_4	FeF_3	CoF_3, CoF_2	NiF_2	CuF_2	ZnF_2	GaF_3	GeF_4	AsF_3, AsF_5	SeF_4, SeF_6	BrF_3	Kr, KrF_2
RbF	SrF_2	YF_3	ZrF_4	NbF_5	MoF_6	TcF_6	RuF_6, RuF_3	RhF_6, RhF_3	PdF_2	AgF, AgF_2	CdF_2	InF_3	SnF_4	SbF_5	TeF_6	IF_5, IF_7	Xe, $XeF_{2,4,6}$
CsF	BaF_2	LaF_3															

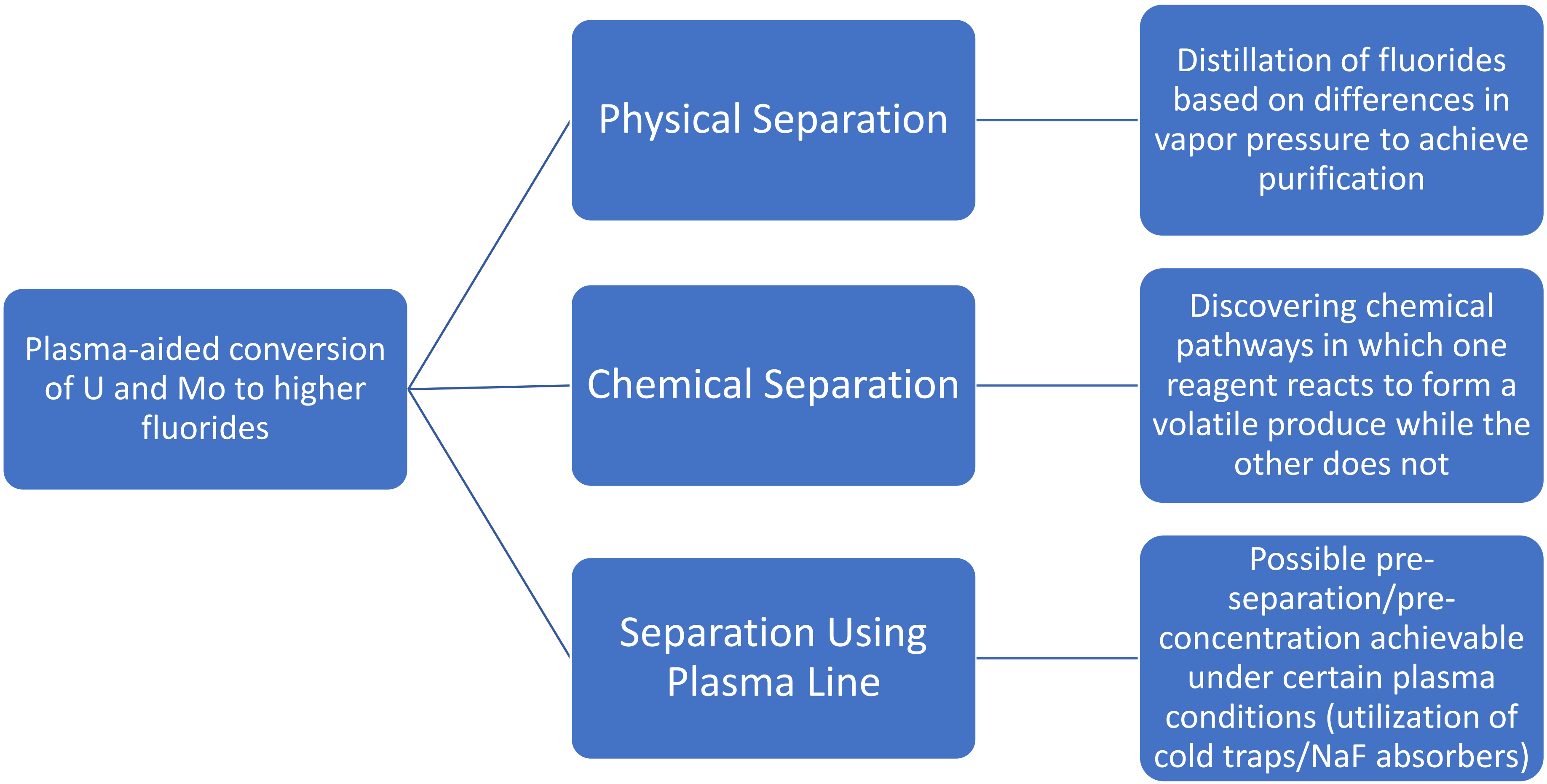
Lanthanoids	CeF_4	PrF_3	NdF_3	PmF_3	SmF_3	EuF_3	GdF_3	TbF_3	DyF_3	HoF_3	ErF_3			
Actinoids	ThF_4	PaF_3	UF_6	NpF_6	PuF_6	AmF_3	CmF_3							

- Major volatile fluorides: UF_6, MoF_6, TeF_6, TcF_6, PuF_6, IF_5, IF_7, NpF_6

- Current focus on the separation of molybdenum from uranium

Proposed Method of Fluoride Separation

- All proposed separation methods are based on dry-chemical techniques. If solvents are considered, only those with very low boiling points will be utilized.



Wet Chemical Processing versus Dry Chemical Processing of Molybdenum-99

	Wet Processing	Dry Processing
Advantages	▪ Produces large quantities of MoO_4^{2-} with high specific activity ▪ Highly developed processes with efficiencies up to 90%	▪ Simple production of targets ▪ Requires processing of only irradiated uranium foil ▪ Significant reduction of liquid radioactive waste ▪ Possible process efficiencies > 90%
Disadvantages	▪ Produces high volumes of liquid radioactive waste	▪ Carrier-free separation yet to be evaluated

Questions?