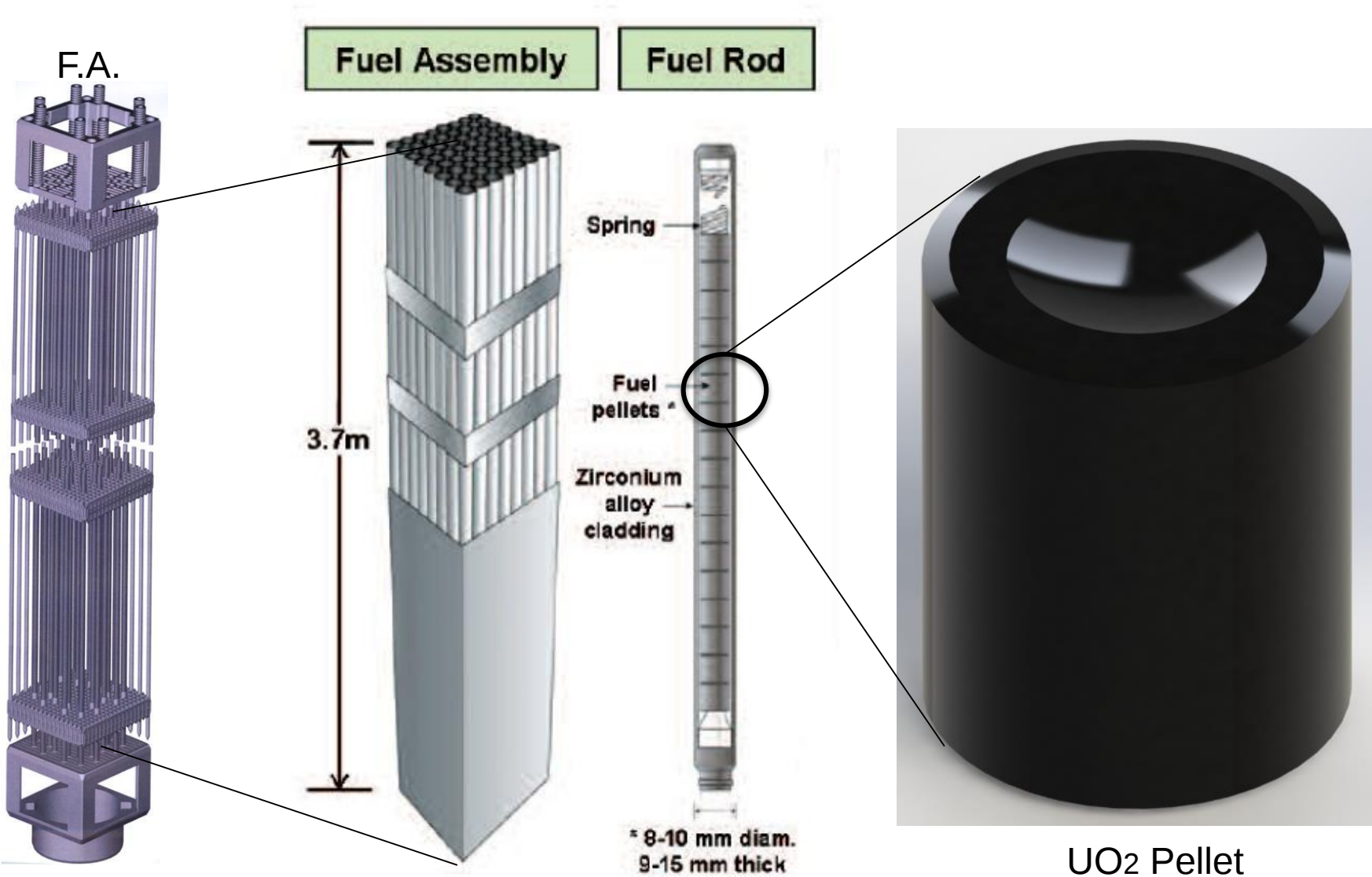


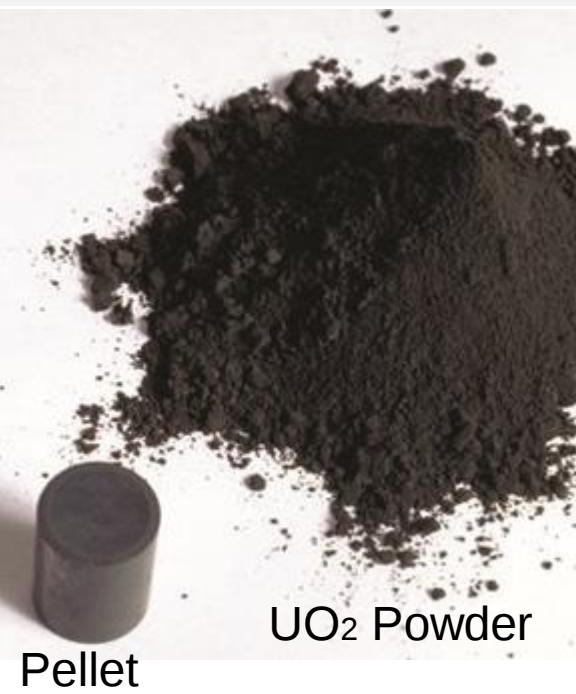
EVALUATION OF THE IMPACTS OF ^{234}U AND ^{235}U HEELS ON THE FORMATION OF A NEW LOT OF COMMERCIAL GRADE UF_6

João P. R. Carnaval¹, Josué C. Medeiros², Davi M. de Oliveira³,
Daniel S. Porfirio⁴, Dalton J. M. C. Beltran⁵ and Bhetina C.
Gomes⁵

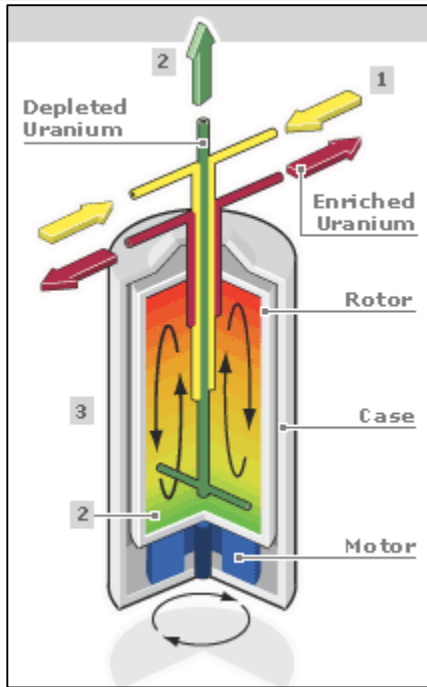
From Fuel Assembly to UO₂ Pellet



From Pellet to UF₆



Uranium Enrichment

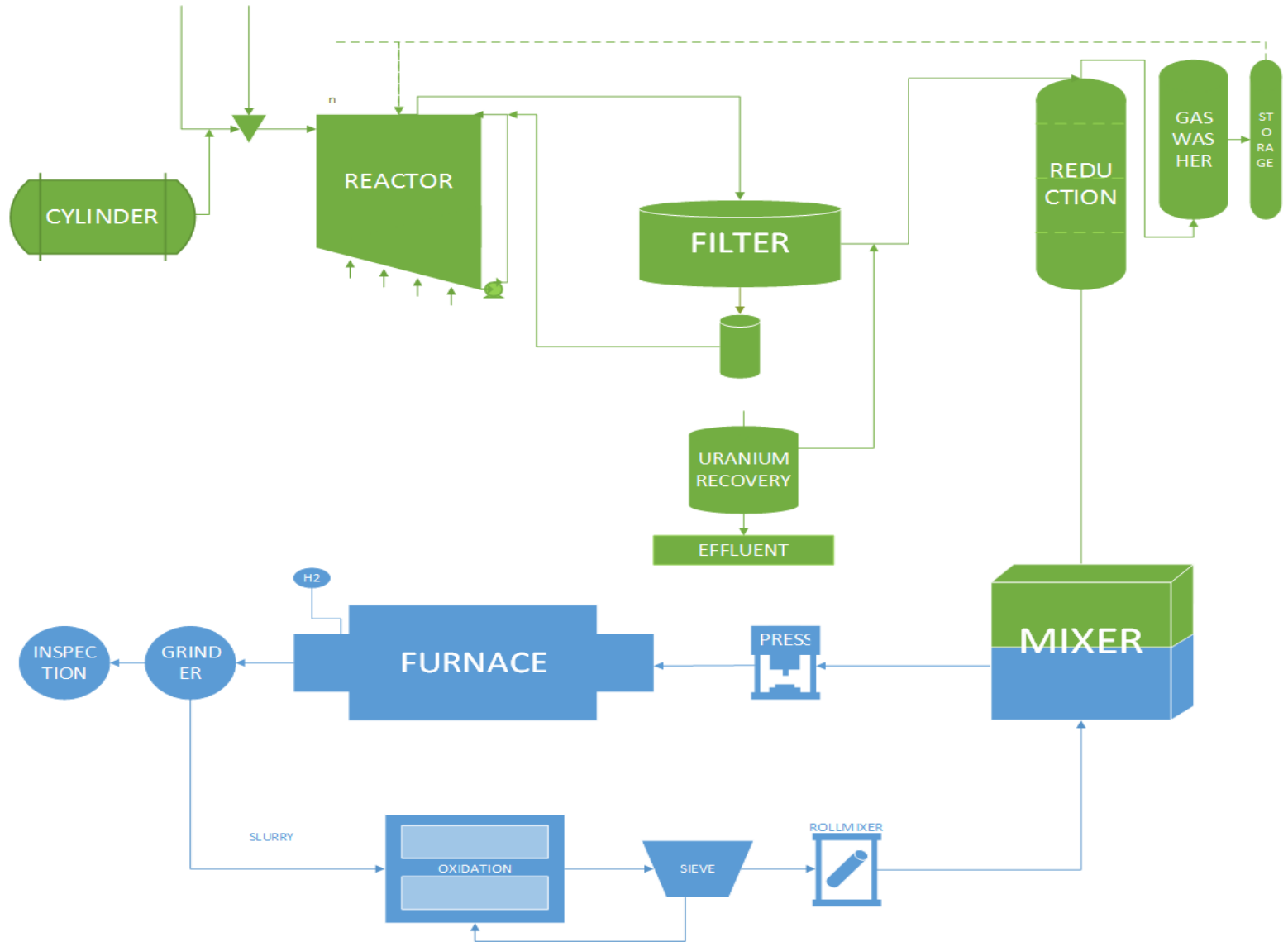


30B cylinder - $<5\%$ ^{235}U

48Y cylinder

48Y cylinder – 0,3% ^{235}U

New Lot of UF₆: Production of AUC



After AUC: Handling the Cylinder with Heel

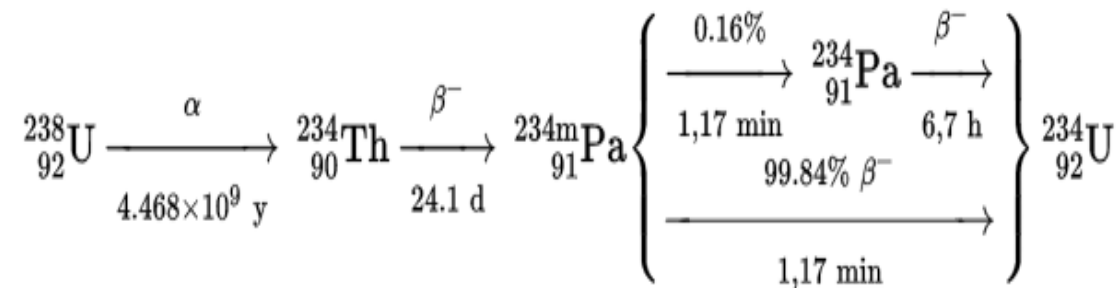


After extraction of UF_6 , the cylinder may contain 11 Kg of UF_6 called *Heel*.

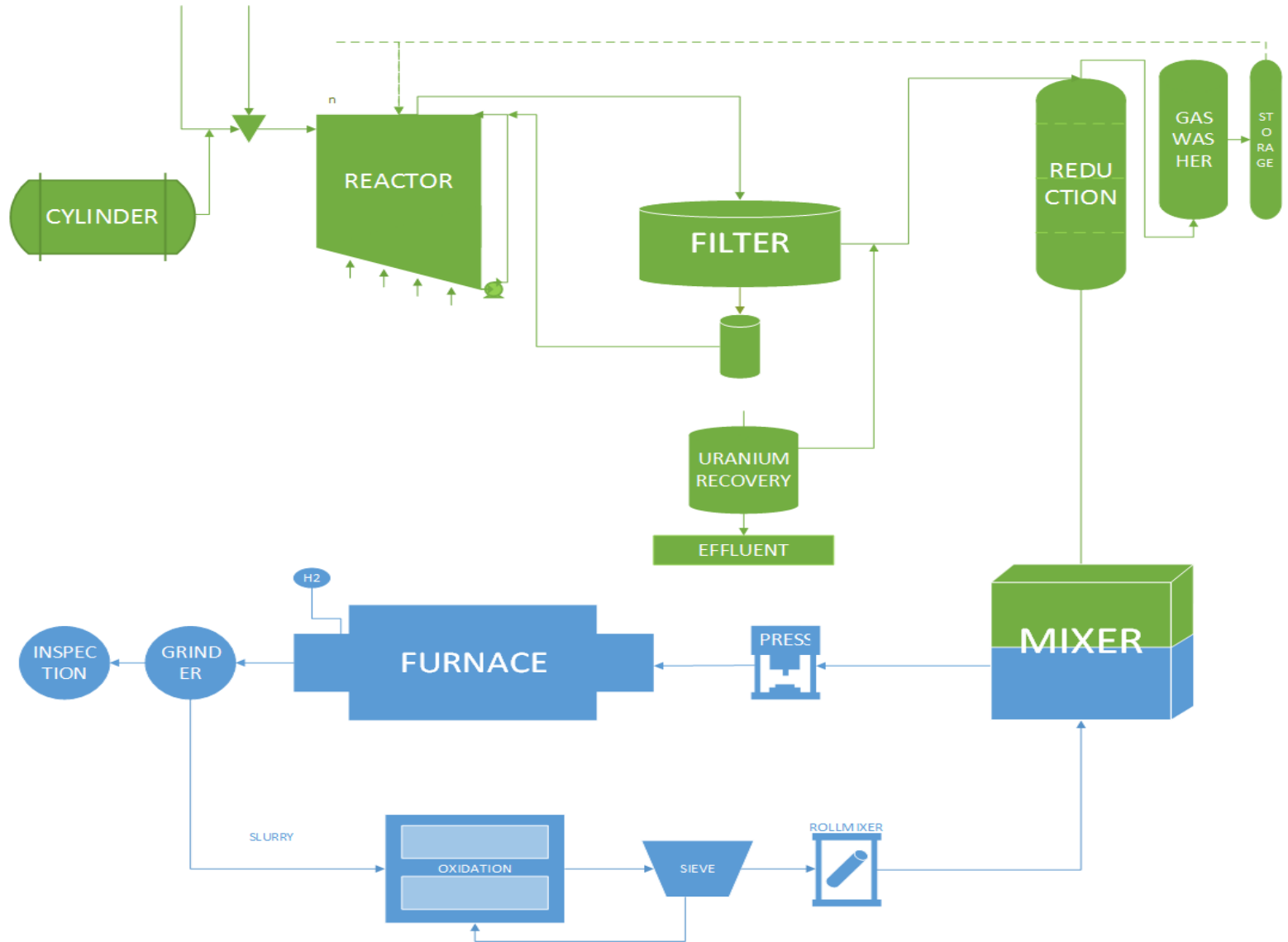
This contains Th and Pa (high emitters), so they must be disposed at an opened area for 6 months until complete decay to ^{234}U according to:

After this period, the options are:

- Reextraction of UF_6
- Washing/Inspection
- Refill with UF_6 for New Lot



New Lot of UF₆: Production of AUC



Formation of a New Lot of UF₆

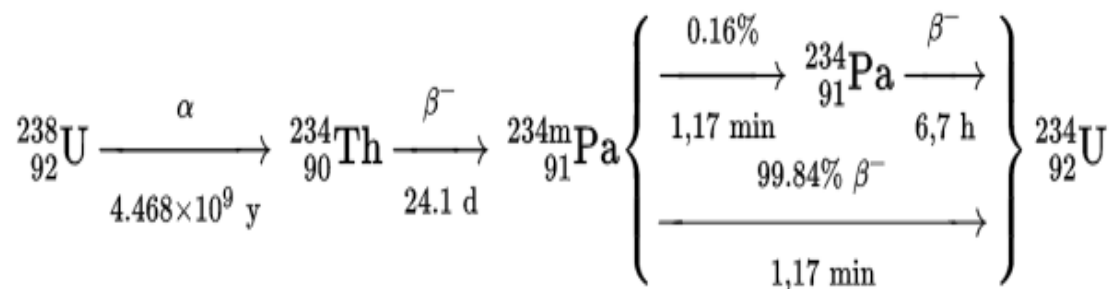
CYLINDER CONDITION

- New Cylinder
- Cleansed Cylinder
- Cylinder containing *heel*

UF6 PROCESS

- Formation of Lot from Isotopic Separation (*IS*)
- Formation of Lot From Mass Transference (*MT*)
 - a. Mass Transference of UF₆ from donor cylinder to new one
 - b. Mass Transference of UF₆ from donor cylinder to clean one
 - c. Mass Transference of UF₆ from donor cylinder to a cylinder containing *heel*
 - d. Mass Transference of UF₆ from donor cylinder to a cylinder containing mass greater than *heel*
- Formation of Lot from Isotopic Recomposition Process (*IR*)

The Study of the Influence of the ^{234}U *Heel* on the lot



$$N_n(t) = \sum_{i=1}^n \left[N_i(0) \times \left(\prod_{j=i}^{n-1} \lambda_j \right) \times \left(\sum_{j=i}^n \left(\frac{e^{-\lambda_j t}}{\prod_{p=i, p \neq j}^n (\lambda_p - \lambda_j)} \right) \right) \right]$$

$$N_D(t) = N_D(t_0) \cdot e^{-\lambda_D \cdot \Delta t} + \lambda_C \cdot N_C(t_0) \cdot \left(\frac{e^{-\lambda_C \cdot \Delta t}}{\lambda_D - \lambda_C} + \frac{e^{-\lambda_D \cdot \Delta t}}{\lambda_C - \lambda_D} \right) + \lambda_B \cdot \lambda_C \cdot N_B(t_0) \cdot \left(\frac{e^{-\lambda_B \cdot \Delta t}}{(\lambda_C - \lambda_B) \cdot (\lambda_D - \lambda_B)} + \frac{e^{-\lambda_C \cdot \Delta t}}{(\lambda_B - \lambda_C) \cdot (\lambda_D - \lambda_C)} + \frac{e^{-\lambda_D \cdot \Delta t}}{(\lambda_B - \lambda_D) \cdot (\lambda_C - \lambda_D)} \right) + \lambda_A \cdot \lambda_B \cdot \lambda_C \cdot N_A(t_0) \cdot \left(\frac{e^{-\lambda_A \cdot \Delta t}}{(\lambda_B - \lambda_A) \cdot (\lambda_C - \lambda_A) \cdot (\lambda_D - \lambda_A)} + \frac{e^{-\lambda_B \cdot \Delta t}}{(\lambda_A - \lambda_B) \cdot (\lambda_C - \lambda_B) \cdot (\lambda_D - \lambda_B)} + \frac{e^{-\lambda_C \cdot \Delta t}}{(\lambda_A - \lambda_C) \cdot (\lambda_B - \lambda_C) \cdot (\lambda_D - \lambda_C)} + \frac{e^{-\lambda_D \cdot \Delta t}}{(\lambda_A - \lambda_D) \cdot (\lambda_B - \lambda_D) \cdot (\lambda_C - \lambda_D)} \right)$$

A = ^{238}U ,
 B = ^{234}Th ,
 C = $^{234\text{m}}\text{Pa}$,
 D = ^{234}U

Calculating the ^{234}U in *Heel*

UF_6	2277	[kg]
Uranium Mass (approximate)	1539.56	[kg]
^{235}U	4.238	[wt%]
^{234}U	8900	[$\mu\text{g/g}^{235}\text{U}$]
^{238}U	95.724	[wt%]

	^{235}U	^{238}U	^{234}Th	$^{234\text{m}}\text{Pa}$	^{234}Pa	^{234}U
Initial Mass [kg]*	65.250	1473.735	0.000	0.000	0.000	0.580
Molar Mass [g/mol]	235	238	234	234	234	234
Half-Life [years]	7.04E+08	4.47E+09	0.065753	2.22603E-06	0.000765	2.45E+05
Decay Constant [s^{-1}]	3.12E-17	4.92E-18	3.33E-07	9.87E-03	2.87E-05	8.99E-14

Period / Type	UF_6 [Kg]	U [Kg]	^{238}U [Kg]	^{235}U [Kg]	^{234}U [Kg]
Day 0-180	2277	1539.5625	1473.5488	65.4314	0.582338791
Day 180-360	11	7.4375	7.1186	0.3161	0.002813234
Day 360	11	7.4375	7.1186	0.3161	0.002813252

For example, 1989 kg of UF_6 (for rounding purposes) at 4.25%wt ^{235}U and containing an initial ^{234}U content of 8900 ppm were added to the existing 11 kg of UF_6 in *heel*. So: ^{234}U Mass increase in the *heel* = $17.67 \mu\text{g}^{234}\text{U}$

Impacts of ^{234}U from Heel in the New Lot

Increase in the ²³⁴ U content on the new lot				Final ²³⁴ U Content on the new lot				
Injected UF ₆ Mass [Kg]	4.25 [%wt ²³⁵ U]	2.6 [%wt ²³⁵ U]	0.85 [%wt ²³⁵ U]	Injected UF ₆ Mass [Kg]	4.25 [%wt ²³⁵ U]	2.6 [%wt ²³⁵ U]	0.85 [%wt ²³⁵ U]	
	²³⁴ U [ppm]				²³⁴ U [ppm]			
2000	0.000306	0.000554	0.001868	2000	8900.0003	8900.0006	8900.0019	
1900	0.000322	0.000577	0.001866	1900	8900.0003	8900.0006	8900.002	
1800	0.00034	0.0006		1800	8900.0003	8900.0006	8900.0021	
1700	0.000359	0.0006		1700	8900.0004	8900.0007	8900.0022	
1600	0.000382	0.0006		1600	8900.0004	8900.0007	8900.0023	
1500	0.000407	0.0007		1500	8900.0004	8900.0007	8900.0025	
1400	0.000436	0.0007		1400	8900.0004	8900.0008	8900.0027	
1300	0.000469	0.0008		1300	8900.0005	8900.0008	8900.0029	
1200	0.000508	0.000919	0.003102	1200	8900.0005	8900.0009	8900.0031	
1100	0.000553	0.001002	0.003381	1100	8900.0006	8900.0010	8900.0034	
1000	0.000608	0.0011		1000	8900.0006	8900.0011	8900.0037	
900	0.000675	0.0012		900	8900.0007	8900.0012	8900.0041	
800	0.000758	0.0013		800	8900.0008	8900.0014	8900.0046	
700	0.000865	0.0015		700	8900.0009	8900.0016	8900.0053	
600	0.001006	0.001822	0.000140	600	8900.001	8900.0018	8900.0061	
500	0.001203	0.002179	0.007351	500	8900.0012	8900.0022	8900.0074	
400	0.001496	0.002709	0.00914	400	8900.0015	8900.0027	8900.0091	
300	0.001977	0.00358	0.012079	300	8900.002	8900.0036	8900.0121	
200	0.002914	0.005276	0.017804	200	8900.0029	8900.0053	8900.0178	
100	0.005539	0.01003	0.033843	100	8900.0055	8900.0100	8900.0338	

ASTM C996

For $^{235}\text{U} < 5\% \text{wt}$

$^{234}\text{U} < 11.000 \text{ ppm}$

Therefore,

Impact of ^{234}U : Null!

Evaluation of the Influence of ^{235}U *heel* in the Processes of the Enrichment Facility

PROCESS OF GENERATION OF UF ₆ TO BE INJECTED IN THE CYLINDER CONTAINING <i>HEEL</i>	CONDITION OF THE <i>HEEL</i> INSIDE CYLINDER TYPE 30B (TO RECEIVE UF ₆ FROM THE PROCESSES)		
	CYLINDER TYPE 30B TO FILL CONTAINING <i>HEEL</i> NO CERTIFICATED (UF ₆ WITH %wt ²³⁵ U UNKNOWN)	CYLINDER TYPE 30B TO FILL CONTAINING <i>HEEL</i> WITH CERTIFICATION (UF ₆ WITH %wt ²³⁵ U KNOWN)	
		<i>HEEL</i> : 0,3 ~ 4,3 %wt ²³⁵ U (±0,05%wt ²³⁵ U)	<i>HEEL</i> WITH ²³⁵ U CONTENT ON NOMINAL VALUE (CLIENT REQUEST)
	IS THE CERTIFICATION REQUIRED FOR THE NEW LOT?		
<i>IS</i>	I. YES	I. YES	I. YES
<i>MT</i>	I. YES	I. MAYBE	I. NO
<i>IR</i>	I. YES	I. MAYBE	I. MAYBE

Applied Example

MASS TO BE TRANSFERED		%wt ²³⁵ U FINAL PRODUCT						
UF ₆ [Kg] 4.25 [%wt ²³⁵ U]	U [g]	<i>Heel 5</i> [%wt ²³⁵ U]	<i>Heel 4</i> [%wt ²³⁵ U]	<i>Heel 3</i> [%wt ²³⁵ U]	<i>Heel 2</i> [%wt ²³⁵ U]	<i>Heel 1</i> [%wt ²³⁵ U]	<i>Heel 0,3</i> [%wt ²³⁵ U]	
2000	1352272.7	INB SPECIFICATION For UF ₆ 4.25%wt ²³⁵ U Tolerance: 4.20 – 4.30%wt ²³⁵ U				38%	4.232%	4.228%
1900	1284659.1					37%	4.231%	4.227%
1800	1217045.5					36%	4.230%	4.226%
1700	1149431.8					36%	4.229%	4.225%
1600	1081818.2					35%	4.228%	4.223%
1500	1014204.5					34%	4.226%	4.221%
1400	946590.9					32%	4.225%	4.219%
1300	878977.3					31%	4.223%	4.217%
1200	811363.6	4.257%	4.248%	4.239%	4.230%	4.220%	4.214%	
1100	743750.0	4.257%	4.248%	4.238%	4.228%	4.218%	4.211%	
1000	676136.4	Therefore ²³⁵ U of Heel affects the lot!				26%	4.215%	4.207%
900	608522.7					23%	4.211%	4.202%
800	540909.1					19%	4.206%	4.196%
700	473295.5					15%	4.200%	4.189%
600	405681.8					09%	4.191%	4.179%
500	338068.2					02%	4.180%	4.165%
400	270454.5					90%	4.163%	4.144%
300	202840.9					70%	4.135%	4.110%
200	135227.3	4.289%	4.237%	4.185%	4.133%	4.081%	4.044%	
100	67613.6	4.324%	4.225%	4.126%	4.027%	3.928%	3.859%	

Conclusion

The effects of ^{234}U and ^{235}U *heels* were evaluated for the production of a new lot of commercial grade UF_6

The Heel with any concentration of ^{234}U does not cause significant effect on the UF_6 lot for any values regarding the specification

The UF_6 *heel* may impact significantly the ^{235}U concentration on the final product and therefore must be controlled in order to remain on the specification limits.

References

ANSI N14.1, “Uranium Hexafluoride – Packing for Transport”, New York, USA, 2012.

M. C. Lee, C. J. Wu, “Conversion of UF_6 to UO_2 : A quasi-optimization of the ammonium uranyl carbonate process,” *Journal of Nuclear Materials* 185, pp.190-201, (1991).

H. Bateman, “Solution of a system of differential equations occurring in the theory of radioactive transformations”, *Proceedings of the Cambridge Philosophical Society*, **vol. 15**, pp. 423–427 (1910).

W.G.P. Auditorium, “Uranium Hexafluoride Handling,” *Second International Conference*, Oak Ridge, Tennessee, (1991).

Material Specification for Commercial Grade Uranium Hexafluoride Enriched to less than 5.000% ^{235}U (weight percent), INB, Resende (2018).

M. Benedict, T. Pigford, H.W. Levi, *Nuclear Chemical Engineering*, McGraw-Hill, New York (1981).

T. Uckan, J. March-Leuba, R. C. Brittain, D. Powell, J. Glaser “Blend down monitoring system fissile mass flow monitor and its implementation at the Siberian chemical enterprise, Springfield, USA (2005). **Vol.44, Nr .3** (1997).

References

- W. Skoog, C. Holler, *Fundamentos da Química Analítica*, Thomson., São Paulo (2006).
- ASTM C996-10, “Standard Specification for Uranium Hexafluoride Enriched to Less 5% ^{235}U ,” Pennsylvania, USA (2011).
- USEC 651, “Uranium Hexafluoride – A Manual of Good Handling Practices”, Ohio, USA (1995).
- Uranium Recovery, Westinghouse Nuclear,
<http://www.westinghousenuclear.com/Portals/0/operating%20plant%20services/fuel/fuel%20products/NFCM-0014%20Uranium%20Recovery.pdf> , (2018).
- D. R. Olander, “The theory of uranium enrichment by the gas centrifuge,” *Progress in Nuclear Energy*, Great Britain, March 1981, **Vol.8**, pp. 1-33 (1981).
- J. Benítez, *Mass Transfer Operation*, John Wiley & Sons, Hoboken, EUA (1948).
- D. C. Stromswold, A. J. Peurrung, P. L. Reeder, D. D. Sunberg, “Neutron methods for monitoring the blending of enriched UF_6 gas”, *IEEE Transactions on Nuclear Science*, **Vol.44**, **Nr .3** (1997).
- W. Skoog, C. Holler, *Fundamentos da Química Analítica*, Thomson., São Paulo (2006).

Thank You
joaocarnaval@inb.gov.br



www.inb.gov.br