WISE Uranium Project - Fact Sheet

Hazards from depleted uranium produced from reprocessed uranium

There has been concern about the detection of uranium-236 in depleted uranium (DU) used for the production of ammunition. U-236 is an artificial nuclide of uranium which only can result from the use of uranium recycled from spent fuel. Therefore, the question is raised, whether other nuclides usually found in spent fuel, such as the transuranics plutonium (Pu-239) and neptunium (Np-237) might also be present in the depleted uranium, and what the health hazard from their presence would be. Due to their heavy atomic weights, transuranics introduced into the enrichment process would concentrate in the tails stream and would therefore show up in the depleted uranium.

The amounts of recycled uranium used in U.S. enrichment plants were first disclosed by the U.S. Department of Energy (DOE) in 1999:

> "At the Paducah uranium enrichment plant, recycled uranium was introduced into the enrichment "cascade" shortly after the startup of the plant in 1953 and continued through 1964. Activities were resumed in 1969 and continued through 1976. Paducah received approximately 100,000 tons (90,000 metric tons) of recycled uranium containing an estimated 328 grams of plutonium, 18.4 kilograms of neptunium and 661 kilograms of technetium-99. Operations at Paducah included the conversion of uranium oxide to uranium hexafluoride at a feed plant located onsite. The converted material was subsequently introduced into the gaseous diffusion "cascade" for further enrichment." [DOE_1999]

For an assessment of the hazards from the transuranics, we first have to determine the concentrations of all nuclides of interest in the depleted uranium. For this purpose, we first need to calculate the mass balance of the enrichment process. We then calculate the inhalation doses from the depleted uranium and compare the dose contributions from the nuclides of interest.

Assay balance for enrichment of reprocessed uranium
(initially enriched to 3.5%, after burnup of 39 GWd/tHM, 5 year storage)

	Feed (a_F)	Product (a_P)	Tails (a_T)
Assay [wt_% U-235]	0.8458	3.82	0.2

[Neghabian_1991] p.90

The ratio of product (enriched uranium) to feed mass is calculated from the assays as follows [Neghabian_1991] p.74:

$$P/F = (a_T - a_F) / (a_T - a_P)$$

Thus, the ratio of tails (depleted uranium) to feed mass is:

$$T/F = 1 - P/F = 1 - (a_T - a_F) / (a_T - a_P)$$

Mass balance for enrichment of reprocessed uranium
(initially enriched to 3.5%, after burnup of 39 GWd/tHM, 5 year storage)

	Feed (F)	Product (P)	Tails (T)
Mass fraction	100.00%	17.84%	82.16%

Concentration of plutonium in tails (depleted uranium) from enrichment of reprocessed uranium, assuming that all plutonium were transfered to the tails:

$$0.328 \text{ kg} / (100{,}000 \text{ st} \cdot 907.185 \text{ kg/st} \cdot 0.8216) = 4.401 \cdot 10^{-9} = 4.401 \text{ ppb}$$

Concentration of neptunium in tails from enrichment of reprocessed uranium uranium, assuming that all neptunium were transfered to the tails:

$$18.4 \text{ kg} / (100{,}000 \text{ st} \cdot 907.185 \text{ kg/st} \cdot 0.8216) = 2.469 \cdot 10^{-7} = 246.9 \text{ ppb}$$

For comparison, we first calculate the inhalation dose from depleted uranium produced from natural uranium. We assume that the short-lived decay products have reached secular equilibrium with their parent nuclides.

Depleted Uranium from Enrichment of Natural Uranium
(from enrichment to 3.5%, tails assay 0.2%)
ICRP72 (public) inhalation, adults, Type S (insoluble forms)

Nuclide	Half-life	Spec. act. [Bq/g]	Conc. [wt_%]	Dose fact. [Sv/Bq]	**Eff. dose** [Sv/g DU]	Dose fraction
U-238	4.468e9 a	1.245e+04	9.980e+01	8.000e-06	9.936e-02	83.73%
Th-234	24.1 d			7.700e-09	9.563e-05	0.08%
Pa-234m	1.17 m					
U-235	7.038e8 a	8.001e+04	2.000e-01	8.500e-06	1.360e-03	1.15%
Th-231	25.52 h			3.300e-10	5.281e-08	0.00%
U-234	2.445e5 a	2.313e+08	8.210e-04	9.400e-06	1.785e-02	15.04%
Total			1.000e+02		**1.187e-01**	100.00%

For depleted uranium from enrichment of reprocessed uranium, the isotope composition is different, and several new nuclides have to be considered - mainly U-236, Pu-239, and Np-237:

Depleted Uranium from Enrichment of Reprocessed Uranium
(initially enriched to 3.5%, after burnup of 39 GWd/tHM, 5 year storage, tails assay 0.2%)
(assuming that all Pu-239 and Np-237 shows up in tails)
ICRP72 (public) inhalation, adults, Type S (insoluble forms)

Nuclide	Half-life	Spec. act. [Bq/g]	Conc. [wt_%]	Dose fact. [Sv/Bq]	**Eff. dose** [Sv/g DU]	Dose fraction
U-238	4.468e9 a	1.245e+04	9.957e+01	8.000e-06	9.913e-02	52.11%
Th-234	24.1 d			7.700e-09	9.542e-05	0.05%
Pa-234m	1.17 m					
U-236	2.342e7 a	2.396e+06	2.266e-01	8.700e-06	4.724e-02	24.83%
U-235	7.038e8 a	8.001e+04	2.000e-01	8.500e-06	1.360e-03	0.72%
Th-231	25.52 h			3.300e-10	5.281e-08	0.00%
U-234	2.445e5 a	2.313e+08	1.939e-03	9.400e-06	4.216e-02	22.16%
Pu-239	24131 a	2.295e+09	4.401e-07	1.600e-05	1.616e-04	0.09%
Np-237	2.14e6 a	2.610e+07	2.469e-05	1.200e-05	7.733e-05	0.04%
Pa-233	27 d			3.900e-09	2.513e-08	0.00%
Total			1.000e+02		**1.902e-01**	100.00%

So, the effective dose from inhalation of this depleted uranium produced from recycled uranium would be 190 mSv/g, compared to 120 mSv/g for DU from natural uranium. This 60% increase would be nearly completely due to the different uranium isotope composition, and only 0.13% of this dose would be caused from the transuranics.

This applies, if all transuranics contained in the reprocessed uranium would have been introduced into the enrichment cascades. This is, however, a gross overestimation:

> "At both Paducah and Oak Ridge sites, the majority of the plutonium and neptunium was separated out as waste during the initial chemical conversion to uranium hexafluoride. Because of this, only a fraction of the plutonium contamination was actually introduced to the gaseous diffusion cascade at either plant. This waste was subsequently reprocessed to recover additional uranium and then reused.
> Of the 328 grams of plutonium present in the 100,000 tons of recycled uranium processed at the Paducah plant, only 0.1 gram of plutonium is estimated to have been introduced into the Paducah cascade. Transuranics including plutonium are believed to have been deposited on internal surfaces of the feed process equipment, with concentrations also being deposited in waste products." [DOE_1999]

According to this estimate, only 0.1 / 328 = 0.03% of the total plutonium would show up in the tails. Therefore, the inhalation dose from plutonium in these tails would cause only 0.09% • 0.03% = 0.000027% of the total dose.

Notes

The uranium isotope concentrations are from [Neghabian_1991] p.90
The concentrations of Pu-239 and Np-237 in reprocessed uranium are from [DOE_1999]; for details see also [DOE_2000].

References

[Neghabian_1991] Verwendung von wiederaufgearbeitetem Uran und von abgereichertem Uran, von A.R. Neghabian, H.J. Becker, A. Baran, H.-W. Binzel, Der Bundesminister für Umwelt, Naturschutz und Reaktorsicherheit (Hg.), Schriftenreihe Reaktorsicherheit und Strahlenschutz, BMU-1992-332, November 1991, 186 S.

[DOE_1999] Past Recycled Uranium Programs Under Review as Energy Department Investigation Continues, U.S. DOE news release R-99-262 of Sept. 29, 1999, http://www.energy.gov/HQPress/releases99/seppr/pr99262.htm

[DOE_2000] Exposure Assessment Project at the Paducah Gaseous Diffusion Plant, Dec. 2000, http://www.eh.doe.gov/benefits/docs/200012paducah.pdf

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