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_1999a]

These figures are based on [DOE_1984]; more details are also available in [DOE_2000].

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.

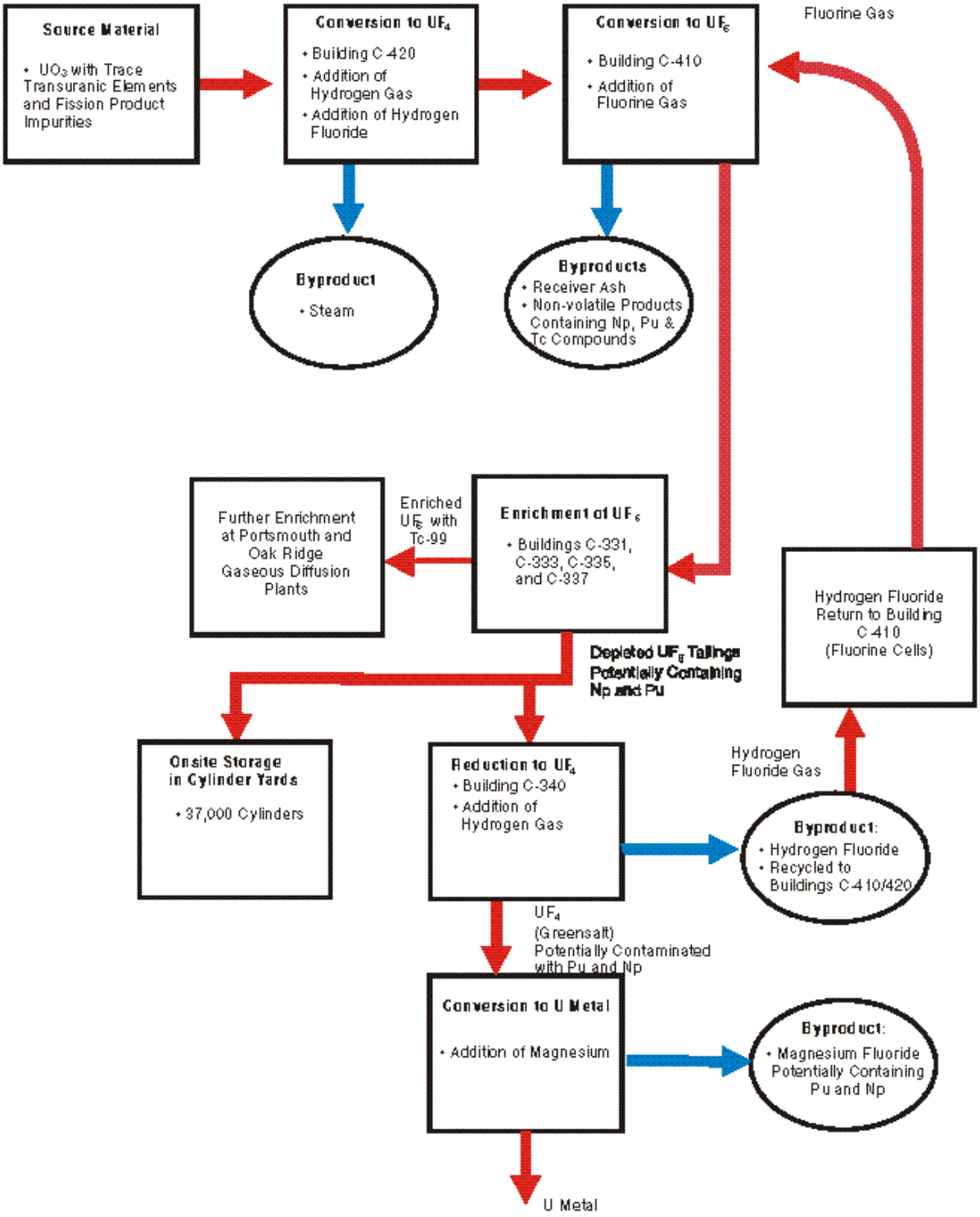
Mass balance for uranium enrichment at Paducah [DOE_1984, p.35]

	Feed	Product	Tails	Other
Mass [st]	758002	124718	621894	11390
Mass fraction	100.00%	16.45%	82.04%	1.50%

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

$$0.328 \text{ kg} / (101{,}268 \text{ st} \bullet 907.185 \text{ kg/st} \bullet 0.8204) = 4.352 \bullet 10^{-9} = 4.352 \text{ ppb}$$

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



Schematic of historic uranium enrichment process at Paducah [DOE_1999b]

$$18.4 \text{ kg} / (101{,}268 \text{ st} \bullet 907.185 \text{ kg/st} \bullet 0.8204) = 2.441 \bullet 10^{-7} = 244.1 \text{ ppb}$$

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 (shown in bold).

Inhalation Dose from 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%

(Nuclide concentrations after [Neghabian_1991])

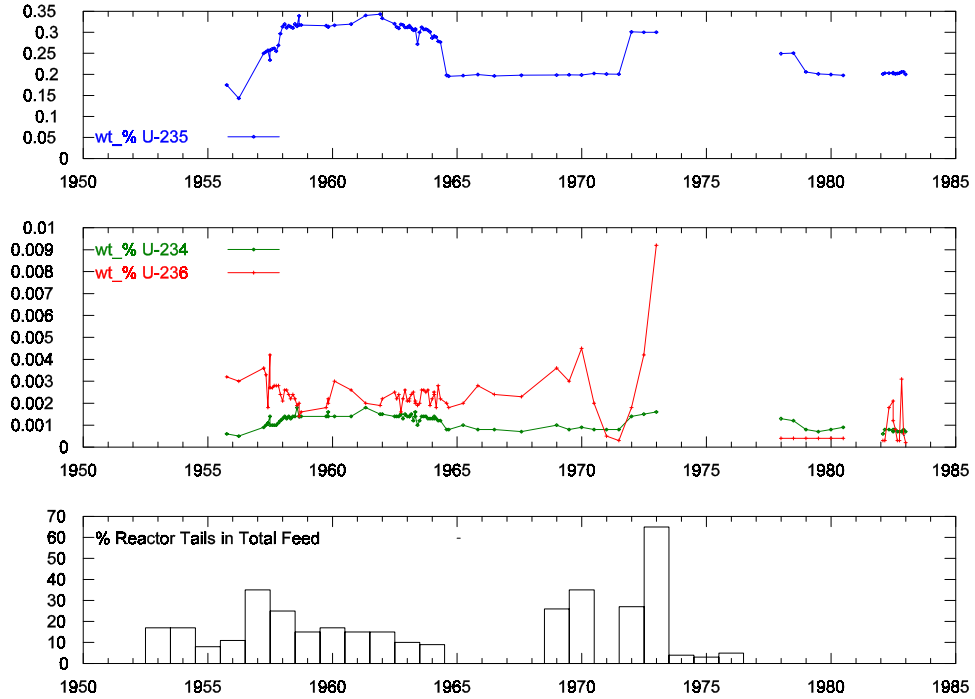
So, the effective dose from inhalation of depleted uranium produced from natural uranium would be 119 mSv/g.

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.

Data from Paducah tails shows concentrations of U-236 of up to 0.0045 wt_%, with typical values in the range of 0.002 - 0.003 wt_% for a tails assay of 0.2% U-235 [DOE_1984 pp.18, 53-55]. Actual monitoring results from DU used for ammunition are as follows: [AEPI_1995] gives a figure of 0.003% U-236; this was confirmed by independent measurements in the U.S. [Dietz_1996]; UNEP found a slightly lower 0.0028% in Kosovo [UNEP_2001a], 0.0027% in Serbia [UNEP_2002a], and 0.0027 - 0.0029% in Bosnia [UNEP_2003a].
Note: these figures are about 75-fold lower than would be expected, if the Paducah feed had been obtained from commercial reactors. This is due the fact that the vast majority of the reprocessed material came from military reactors in Hanford and Savannah River (low burnup), and that the reprocessed material constituted only approx. 13% of the Paducah feed.

U-234 concentrations in Paducah tails ranged rom 0.0006 to 0.0010 wt_% for a tails assay of 0.2% U-235 [DOE_1984 p.15]; this is a typical tails assay for the DU used in ammunition [AEPI_1995].

Minor uranium isotopes in Paducah tails [after DOE_1984]



With this data, we obtain the following results:

Inhalation Dose from Depleted Uranium used in DU penetrators
(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	9.980e+01	8.000e-06	9.936e-02	83.45%
Th-234	24.1 d			7.700e-09	9.563e-05	0.08%
Pa-234m	1.17 m					
U-236	2.342e7 a	2.396e+06	3.000e-03	8.700e-06	6.254e-04	0.53%
U-235	7.038e8 a	8.001e+04	2.000e-01	8.500e-06	1.360e-03	1.14%
Th-231	25.52 h			3.300e-10	5.281e-08	0.00%
U-234	2.445e5 a	2.313e+08	8.000e-04	9.400e-06	1.739e-02	14.61%
Pu-239	24131 a	2.295e+09	4.352e-07	1.600e-05	1.598e-04	0.13%
Np-237	2.14e6 a	2.610e+07	2.441e-05	1.200e-05	7.645e-05	0.06%
Pa-233	27 d			3.900e-09	2.485e-08	0.00%
Total			1.000e+02		**1.191e-01**	100.00%

So, the inhalation dose from DU used for penetrators would be only 0.7% higher than from DU obtained from enrichment of natural uranium. U-236 would contribute 0.53% to the dose, Pu-239 0.13%, and Np-237 0.06%.

The above calculations have assumed that 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_1999a] (emphasis added)

If it is assumed, that all of these 0.1 g of plutonium were transfered to the tails, the plutonium-concentration in the tails would be 0.0013 ppb. It is, however, questionable whether any significant fraction of these 0.1 g of plutonium was transfered to the tails, according to DOE [DOE_1984 p.17].
There exists only sporadic monitoring data of plutonium concentrations in Paducah tails and in DU metal made from it for the years the reprocessed uranium was fed into the cascade. In no case has plutonium been found in amounts above the detection limit given by the respective measuring techniques used:

Monitoring data for plutonium in Paducah tails and products made thereof

Year	Item	Plutonium concentration	Reference
1957	DU metal	< 1 ppb (based on U)	[DOE_1963]
1963	tails	< 1 ppb (based on U)	
1964	tails	< 10 ppb	[DOE_1984, p.17]
1973	tails	< 0.01 ppb	
from 1975	tails	< 0.01 ppb	

1 ppb = 1 part per billion = 10^{-9}

And, of the18.4 kg of Np-237, only 4.6 kg is estimated to have been fed into the cascade. [DOE_1984 p.11]

According to these estimates, only less than 0.03% of the total plutonium and 25% of the total neptunium could have shown up in the tails. **Therefore, the inhalation dose from plutonium would cause only less than 0.000039% of the total dose, and the dose from neptunium would cause less than 0.016% of the total dose from the DU used for penetrators.**

Since February 2001, first monitoring results for plutonium in DU penetrators spent in Kosovo

are available. In several cases, the detection limit was low enough to actually find traces of plutonium. The results confirm, albeit for a few penetrators only, that the above assumptions (0.0013 ppb) are realistic. New data from penetrators recovered from target areas in Southern Serbia and Bosnia-Herzegovina shows plutonium concentrations up to 30 times higher.

Monitoring data for plutonium in uranium penetrators

Location	Plutonium concentration	Reference
Kosovo	< 0.0032 ppb (based on U)	[GSF_2001]
Kosovo	0.00035 - 0.0056 ppb	[UNEP_2001b]
Southern Serbia	0.019 ppb	[McLaughlin_2003]
Southern Serbia	0.0058 - 0.0138 ppb	[UNEP_2002a]
Bosnia	0.0022 - 0.0382 ppb	[UNEP_2003a]

1 ppb = 1 part per billion = 10^{-9}

For comparison: plutonium concentrations in the range of a few thousandth parts of a ppb are naturally found in uranium ore deposits: uranium-238 captures neutrons coming from various natural sources, such as cosmic radiation, and spontaneous fission of uranium-235. The product is uranium-239, which decays at a half-life of 23.4 minutes to neptunium-239, which, in turn, decays at a half-life of 2.355 days to plutonium-239. The plutonium actually found in penetrators would, however, nearly completely be from artificial sources. This is a result of the chemical processing of the material, reducing plutonium concentrations from any source.

In its 2003 assessment for Bosnia-Herzegovina, UNEP also reports concentrations of neptunium for 3 penetrators recovered:

Monitoring data for neptunium in uranium penetrators

Location	Neptunium-237 conc.	Reference
Bosnia	< 0.15 - 0.62 ppb	[UNEP_2003a]

1 ppb = 1 part per billion = 10^{-9}

The only database available so far of more than sporadic monitoring data of contaminants found in DU is for DU metal used for the fabrication of tank armor: The Idaho Nuclear Technology and Engineering Center (INTEC) has analyzed 60 samples of depleted uranium metal billets for transuranics and fission products [Army_2000]. Transuranics concentrations above the detection limits have been identified in this material, including not only plutonium-239, but also americium-241, neptunium-237, and plutonium-238. Furthermore, the fission product technetium-99 was detected.

Monitoring data for transuranics and fission products in DU armor

	Nuclide data			max. values found in DU armor		
	Half-life	Specific Activity [Bq/g]	Dose Coeff. [3]) [Sv/Bq]	Activity Conc. [2]) [Bq/g DU armor]	Conc. by weight [ppb]	Effective Dose [3]) [Sv/g DU armor]
Am-241	432.2 a	1.271e+11	4.2e-05	0.703	0.0055	3.0e-05
Np-237	2.140e6 a	2.611e+07	2.3e-05	0.137	5.2470	3.2e-06
Pu-238	87.75 a	6.340e+11	4.6e-05	0.074	0.0001	3.4e-06
Pu-239/240 [1])	24.13e3 a	2.296e+09	5.0e-05	0.1	0.0436	5.0e-06
Tc-99	213.0e3 a	6.280e+08	4.0e-08	19.98	31.8153	8.0e-07
Total						4.2e-05

[1]) nuclide data of Pu-239 only
[2]) [Army_2000]
[3]) ICRP 72 (Public) Inhalation, Adults, AMAD = 1 μm, Class M (Note: Other than for uranium and technetium, the inhalation dose coefficients for the transuranics increase rather than decrease with the solubility of the material.)

Thus, for DU armor containing 0.2% U-235 and 0.003% U-236, **the excess inhalation dose from transuranics and fission products of max. 0.042 mSv/g represents only a 0.035% increase over the dose from the DU alone**.

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WISE Uranium Project, Peter Diehl
http://www.wise-uranium.org

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