Depleted and Recyclable Uranium in the United States: Inventories and Options

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International consumption of uranium currently outpaces production by nearly a factor of two. Secondary supplies from dismantled nuclear weapons, along with civilian and governmental stockpiles, are being used to make up the difference but supplies are limited. Large amounts of $^{235}$U are contained in spent nuclear fuel as well as in the tails left over from past uranium enrichment. The usability of these inhomogeneous uranium supplies depends on their isotopes. We present data on the $^{235}$U content of spent nuclear fuel and depleted uranium tails in the US and discuss the factors that affect its marketability and alternative uses.

I. INTRODUCTION

Historically, the majority of commercial nuclear power has been generated using light-water reactors (LWRs) burning low enriched uranium. While research into technologies that could close the fuel cycle continues within the Global Nuclear Energy Partnership (GNEP) and elsewhere, the maturity and economic competitiveness of LWRs will make them a major presence for decades to come. Because of this, global demand for uranium is likely to remain strong and its future price uncertain, with acceptable alternatives to mined natural uranium being of significant interest as a result. At present, substitutes include down-blending of highly enriched uranium, uranium released from government or utility stockpiles, enrichable depleted uranium (DU) and reprocessable uranium (RU) from spent LWR fuel (SF).

II. INVENTORIES

The decision of whether to mine fresh uranium or exploit alternative sources is largely a matter of economics. Both RU and DU stockpiles have a variable $^{235}$U composition and will often require additional enrichment beyond what is needed for manufacturing LWR fuel from natural uranium, particularly in the case of RU because of $^{236}$U poisoning. As a result, the price of using DU will depend on the costs of enrichment, DU cylinder transport from storage to the enrichment plant, $\text{UF}_6$ tails storage, deconversion of $\text{UF}_6$ tails to $\text{U}_3\text{O}_8$ and its subsequent disposal. Using RU will have the additional costs of spent fuel transportation, reprocessing and waste disposal. In both cases the total life cycle costs for using RU and DU for making LWR fuel should be compared to those of disposal, for which the US government is responsible.

II.A. Depleted Uranium

Inventories of DU, in the form of uranium hexafluoride ($\text{UF}_6$), have been accumulating since the beginning of the nuclear age and the US currently holds 708,189 tonnes of $\text{UF}_6$ in 58,890 cylinders stockpiled at Paducah, Kentucky and Portsmouth, Ohio.

Table I shows the amount of depleted uranium in $\text{UF}_6$ form in the US as a function of $^{235}$U assay. The $\text{UF}_6$ is stored in 58,890 cylinders at Paducah and Portsmouth. In total, as of the end of 2006, there are 708,189 million tonnes of $\text{UF}_6$ in the US. Data are from the DOE Office of Nuclear Management Information Database, December 2006.

<table>
<thead>
<tr>
<th>Assay range ($^{235}$U)</th>
<th>tonnes $\text{UF}_6$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1250 - 0.1649</td>
<td>149</td>
</tr>
<tr>
<td>0.1650 - 0.2149</td>
<td>174137</td>
</tr>
<tr>
<td>0.2150 - 0.2649</td>
<td>192883</td>
</tr>
<tr>
<td>0.2650 - 0.3149</td>
<td>135056</td>
</tr>
<tr>
<td>0.3150 - 0.3649</td>
<td>151952</td>
</tr>
<tr>
<td>0.3650 - 0.4149</td>
<td>23989</td>
</tr>
<tr>
<td>0.4150 - 0.4649</td>
<td>10535</td>
</tr>
<tr>
<td>0.4650 - 0.5149</td>
<td>425</td>
</tr>
<tr>
<td>0.5150 - 0.5649</td>
<td>1163</td>
</tr>
<tr>
<td>0.5650 - 0.6149</td>
<td>94</td>
</tr>
<tr>
<td>0.6150 - 0.6649</td>
<td>227</td>
</tr>
<tr>
<td>0.6650 - 0.7149</td>
<td>17580</td>
</tr>
</tbody>
</table>

The total inventory of uranium declared surplus by DOE – 59.2 million kg of natural $\text{U}_3\text{O}_8$ equivalent as of May 2006, includes HEU and NU as well as DU [1]. Of this excess uranium, 24.5 million kg is HEU to be blended to LEU; most of the rest is NUF$_6$ or DUF$_6$ “of economic value.” To avoid distorting effects that would accompany large-scale dumping, DOE proposes to place on the market no more than 10% of the annual fuel requirements of the domestic reactor fleet, or about 5 million tonnes natural uranium equivalent per year.
The DU listed in Table I contains the same number of
$^{235}$U atoms as 180,000 tonnes of natural uranium. Simple
mass balance calculations show that if all of this DU were
enriched to 4.2% with tails assay of 0.1%, over 26,000
tones of reactor fuel – about 12 years’ supply at current
consumption rates – could be produced. To enrich all
US-held DU to 4.2%, though, would require about 500
million SWU, or 44 years worth of full-time deployment
of the current US enrichment capacity of 11.3 million
SWU/year. Therefore, it is likely that the pace of DU re-
enrichment will be set by the amount of excess SWU
capacity available at any given time, its price and that of
NU. This will be discussed in further detail in the section
on Utilization Options.

Adjustment of DU tails assay is the sole short-term
method of introducing demand elasticity available to
utilities. Prior to 2000, the prevailing DU tails assay in the
United States was 0.3 w/o $^{235}$U. As the price of uranium
has increased, the front-end cost-minimizing tails assay
has decreased to less than 0.2 w/o $^{235}$U, although even as
NU prices have risen the multi-year nature of many
separate work contracts has caused actual tails assays to
lag the cost-minimizing value for utilities. To place this in
context, when producing 4.2% enriched fuel a reduction
of tails assay from 0.3 to 0.2 w/o will decrease natural
uranium requirements by 18%. Hence, market forces can
lead to considerable economies of primary uranium
consumption.

DU will of course continue to accumulate, although it
is difficult to project the average assay of future holdings.
For purposes of illustration we will assume an average
future assay of 0.25%, reflecting assays that had been
prevailing in Europe for decades. The U.S. Department of
Energy currently plans to deconvert the majority of
stockpiled UF$_6$ to U$_3$O$_8$ for stable storage until final
disposal at a deconversion cost of $2.80 /kg UF$_6$.[2].
In the Utilization Options section the relative merits of
immobilization versus storage in hexafluoride form –
preserving the option to re-enrich – are discussed in the
context of both current DU holdings and future
accumulations.

II.B. Reprocessable Uranium

Holdings of DU and RU in the United States contain
roughly equal quantities of $^{235}$U. While DU inventories
are already characterized by isotopic content, analysis was
needed to obtain the equivalent data for RU. RU
isotopes are most strongly dependent on the initial
enrichment of the fuel and its subsequent discharge
bunup. We obtained data on SF discharge dates, bunup
and reactor prevalence (PWR vs. BWR) from The
Nuclear Fuel Data Survey [3]. We subsequently
conducted bunup calculations using ORIGEN 2.2 [4] in
which discharge bunup and initial enrichment were
parametrically varied over the applicable range (15 to 55
MWD/kgHM bunups and initial enrichments in the
range of 2.0 to 4.4 w/o $^{235}$U).

To characterize the legacy SF in the United States
both the SF inventory and its composition are needed.
The mass distribution of the legacy SF inventory, in terms
of each of its component isotopes, fully characterizes the
legacy fuel. It is a challenging task to determine this
distribution because the initial enrichments and bunup
histories of the legacy SF assemblies are quite diverse.

Historical data concerning the United States SF
inventory was obtained from the Energy Information
Administration (EIA). The EIA maintains records on all
fuel assemblies in the United States and includes data on
every fuel assembly’s mass, bunup, initial enrichment,
and the reactor in which it was irradiated. At present the
EIA has these data on 29,454 spent nuclear fuel
assemblies which is current through 2002.

ORIGEN2.2 bunup calculations offer a simple,
medium-fidelity route toward characterizing the SF
compositions. To specify the irradiation times and power
densities required to carry out an ORIGEN2.2 bunup
calculation, a general model of typical irradiation cycles
was developed.

Since the United States fleet contains boiling water
reactors (BWRs) and pressurized water reactors (PWRs)
nominal values for the fueling cycles of these two were
taken. In addition, PWRs and BWRs have sufficiently
distinct neutron spectra to warrant separate treatment even
to meet the relatively low-fidelity needs of this
application. For BWRs, only one cross section library
was used in performing bunup calculations. This library,
calculated assuming a discharge bunup of 33 MWD/kg,
was used for the entire range of bunups for BWRs of 15
– 50 MWD/kg. For PWR fuel, which constitutes the bulk
of US SF, characteristic cross section libraries existed (or
were prepared) for 33, 50, and 60 MWD/kg discharge
bunups. When a bunup fell between two of the values
for which libraries existed, interpolation was used to form
a new library specific to that bunup. If the bunup was
higher or lower than any of the relevant libraries, the
highest or lowest cross section library was used
respectively. For PWRs the bunups ran from 15 – 70
MWD/kg. For both types of reactors, the enrichment
varied from 2% to 4.4%.

By varying both the initial enrichment and bunup for
fuel each reactor type, spent fuel isotopic compositions
can be calculated for each assembly with ORIGEN2.2.
However given the large number of assemblies and the
relatively small set of parameters needed to describe the
burnup history of an assembly, an algorithm for creating a look up table of SF compositions was developed.

Given that typical reactor power densities, batchwise fuel management schemes and load factors are assumed, only enrichment and discharge burnup remain as variables distinguishing the composition of the spent fuel assemblies (post-irradiation decay is handled separately and is discussed below). For a given burnup and reactor type, the enrichment was determined in steps of size 0.1% for the whole range from 2-4.4%. For each of these enrichments and burnups, the discharge composition of the respective SF was found. This was then repeated for all burnups in their range (15-55 MWD/kg for BWR, 15-70 MWD/kg for PWR) using a step size of 2 MWD/kg.

\[
u(BU,e_o) = k\cdot\exp\left[-(A\cdot BU + B\cdot e_o)\right] \quad (1)
\]

Here \(\nu\) is the weight percent of initial heavy metal of a given isotope at discharge with \(A, B\) and \(k\) being fitting parameters. \(A\) has units of \(\text{kg/MWD}\), \(B\) of (weight percent of initial heavy metal at charge)*, and \(k\) of weight percent or initial heavy metal at discharge. The variables \(BU\) and \(e_o\) represent the burnup and enrichment respectively. Once the specific parameters \(A, B,\) and \(k\) have been found, the data from the EIA database can be used in conjunction with Equation 1 to find the uranium composition for all fuel assemblies.

Equation 1 and Table II depict the method used to determine the uranium isotopic concentrations in SF. Although each uranium isotope was subjected to this analysis, as were isotopes such as \(^{238}\text{Pu}\) that are decay parents of uranium isotopes, only the results for \(^{235}\text{U}\) and \(^{236}\text{U}\) will be presented here.

**TABLE II. Parameters in Equation 1**

<table>
<thead>
<tr>
<th>(k)</th>
<th>(A)</th>
<th>(B)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.670</td>
<td>0.0448</td>
<td>-0.509</td>
</tr>
<tr>
<td>0.640</td>
<td>0.0392</td>
<td>-0.497</td>
</tr>
<tr>
<td>0.106</td>
<td>-0.0124</td>
<td>-0.264</td>
</tr>
<tr>
<td>0.105</td>
<td>-0.0129</td>
<td>-0.259</td>
</tr>
</tbody>
</table>

Since the EIA database gives the mass of each fuel assembly, the uranium isotopic vectors can easily be combined with the assembly masses to compute their total uranium content. It must be recognized that the EIA database gives the burnup of a fuel assembly only as an average over the whole assembly. Since the isotopic compositions do not vary linearly with burnup, the true spatial power distribution would be needed to increase the fidelity of this calculation. Since each reactor and fuel loading is characterized by a unique flux profile, there is no elementary way to take this complexity into account. Indeed, the extra information needed to resolve assembly burnup profiles beyond the given averages may be lost to history along with the records of the detailed fuel loading patterns to which the assemblies were subjected. However, the fidelity of using one group ORIGEN2.2 cross sections is a more weighty concern. Although these cross sections were allowed to vary with burnup to the extent supported by ORIGEN2.2, a higher-fidelity (and more time-consuming) approach would populate the lookup tables via - at the least - pin-cell level transport-burnup calculations. Such an option would be strongly desirable for repository impact calculations, for instance,

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Fig. 1. Structure of Lookup Table for SF Uranium Characterization

In a more algorithmic sense, this process contains three nested loops. The top loop is the reactor type, the middle loop is the burnup, and the bottom loop is the enrichment of \(^{235}\text{U}\). All of this serves to create a set of 3-tuple data set for each reactor type. The first coordinate is the burnup, the second is the enrichment, and the third is a vector of the isotopic composition of SF. A schematic of the lookup table thus produced - using very coarse bins for compactness of illustration - appears as Figure 1. The \(u_i\) are individual uranium isotopic vectors at discharge associated with the midpoint values of a given (enrichment, burnup) bin.

The above method was used with ORIGEN2.2 to populate lookup tables, and the results were used in combination with the EIA data to generate data on the uranium compositions of the current US SF stockpile. The complete data set for each fuel assembly type was fitted to the equation:

\[
u(BU,e_o) = k\cdot\exp\left[-(A\cdot BU + B\cdot e_o)\right] \quad (1)
\]
but it was deemed unnecessary for cost-benefit analysis purposes.

The calculated cumulative distribution of uranium in US legacy SF by its $^{235}$U content is shown in Figure 2. It can be seen from the figure that the median $^{235}$U enrichment of RU is about 0.85%. It is important to note that each residual $^{235}$U enrichment level is associated with some spread of $^{236}$U assay, which is shown in Figures 3 and 4. Figure 3 shows the normalized cumulative distribution of $^{236}$U in uranium that contains 0.55-0.65% $^{235}$U when it comes out of the reactor. Figure 4 shows the same normalized distribution for RU containing 0.85-0.95% $^{236}$U. Figures 3 and 4 are normalized for convenience of comparison; in fact, there is a great deal more uranium in the 0.85-0.95% $^{235}$U bin than in the 0.55-0.65% bin.

Fig. 2. Estimated $^{235}$U Content of Uranium Contained in US SF.

Fig. 3. Cumulative distribution function of estimated normalized $^{236}$U content of RU with 0.55-0.65% $^{235}$U. The ordinate shows the fraction of RU that has a $^{236}$U content less than or equal to the value given on the abscissa.

Fig. 4. Cumulative distribution function of estimated normalized $^{236}$U content of RU with 0.85-0.95% $^{235}$U. The ordinate shows the fraction of RU that has a $^{236}$U content less than or equal to the value given on the abscissa.

Table III gives the percentage of the 46,999 tonnes of uranium contained, as of December 31 2002, in US SF as a function of $^{235}$U assay. Complete actinide isotopes have been compiled for each SF assembly, though Table III only shows $^{235}$U composition.

### TABLE III. Calculated Assay Distribution of US Recyclable Uranium

<table>
<thead>
<tr>
<th>$^{235}$U Assay Range [%]</th>
<th>Percentage of US RU</th>
</tr>
</thead>
<tbody>
<tr>
<td>&lt; 0.55</td>
<td>2.35</td>
</tr>
<tr>
<td>0.55-0.65</td>
<td>10.03</td>
</tr>
<tr>
<td>0.65-0.75</td>
<td>22.22</td>
</tr>
<tr>
<td>0.75-0.85</td>
<td>27.20</td>
</tr>
<tr>
<td>0.85-0.95</td>
<td>19.74</td>
</tr>
<tr>
<td>0.95-1.05</td>
<td>8.63</td>
</tr>
<tr>
<td>1.05-1.15</td>
<td>3.64</td>
</tr>
<tr>
<td>1.15-1.25</td>
<td>1.80</td>
</tr>
<tr>
<td>1.25-1.35</td>
<td>1.24</td>
</tr>
<tr>
<td>1.35-1.45</td>
<td>0.63</td>
</tr>
<tr>
<td>&gt; 1.45</td>
<td>2.52</td>
</tr>
</tbody>
</table>

### III. OPTIONS AND FUTURE WORK

Within the context of the GNEP only a handful of uses for DU exist beyond re-enrichment. Depleted uranium can realistically be used as a matrix for downblending highly enriched uranium from dismantled nuclear weapons, in energy storage fly wheels, or for use as fast reactor blanket material. However, only its use in shielding applications for spent fuel storage casks appears capable of absorbing the amount of material that is
currently stockpiled. This disparity is likely to grow with time, especially if demand for nuclear power increases.

The US Department of Energy currently plans to deconvert the UF₆ to U₃O₈ for stable storage until final disposal, the form of which is still open to debate. This path is favored in part because only a limited number of uses for DU and RU exist beyond re-enrichment, which has itself been more expensive historically than mining fresh uranium. Nonetheless, given the sharp rise in primary uranium prices, it has become important to identify scenarios in which DU and RU could act (for a period of time) as cost-effective partial alternatives to further uranium mining. The options to be considered for DU are listed below.

Current DOE strategy:
- Conversion of UF₆ to U₃O₈ with subsequent disposal
  i) Conversion of UF₆ to U₃O₈ costs $ 2.80/kg UF₆ [2]
  ii) Disposal cost estimates for U₃O₈ range from $5-110/kg [5]

Alternate Strategies:
A. Domestic Recenrichment of UF₆
   i) to 0.71% or 1%, using excess separative work capacity, followed by storage as UF₆
   ii) keep storing as UF₆ until reenrichment is called for

B. Shipment of UF₆ Abroad, Recenrichment and Return to the US
   i) same options as above, however considerable additional excess capacity exists abroad particularly in Russia
   ii) there is a precedent: Germany, France and, very recently, Japan have pursued an arrangement of this type with Russia
   iii) a significant technical obstacle to realization of this strategy exists: the transport of UF₆ casks is subject to complex regulations; many casks could not be transported abroad and others would require overpacks

C. Conversion of UF₆ to U metal for use in spent fuel disposal casks
   i) cask cost has been estimated to be $22.80/kg UF₆ in 1994 dollars [6]
   ii) this strategy would immobilize nearly all current DU holdings. However, the fate of DU produced in the future would still be open to question

D. Conversion of UF₆ to UOX or metal for use as matrix in MOX, Fast burner reactor fuel, or for downblending HEU
   i) Even under aggressive MOX fuel or burner reactor deployment plans, system studies [7] have shown that only a few percent of DU produced each year could be recycled in this way. The same is true for downblending of HEU

II. Non-nuclear: e.g., conversion of UF₆ to U metal for use in fly wheels
i) Non-nuclear military applications include use in ballistics and armor. The use of DU in energy storage flywheels or counterweights has also been explored [5,6]. However, the amount of material required to meet these applications is small compared to the current supply.

Options for RU beyond re-enrichment are even more proscribed. Moreover, disposal of RU is fraught with greater political concerns and constraints than is the case for DU. Since re-enrichment of RU will still require the disposal of radioactive tails, the use of RU in fast reactor blankets could be an attractive option. Once again, though, unless a breeding fuel cycle comes to fruition, RU recycle as a matrix material in MOX or fast reactors would be insufficient to immobilize more than a few percent of the RU produced annually by the LWR fleet.

Although commercial RU enrichment is not presently taking place (subject to change in the near term in view of Russian-Japanese negotiations), the U.S., Russia and France have enriched RU in significant quantities in the past. The most recent major study of RU enrichment in the U.S. took place in 1993 [8]. Although this study must now be considered outdated, it is certain that the most substantial additional expenses that adhere to RU enrichment are purification and tails disposal. Both of these issues are complicated by the presence of the isotope ²³²U. Although ²³²U (T½ = 68.9 yr) is present in RU at levels of a few parts per 10¹⁵ atoms, ²³⁶Th and other daughters in its chain that undergo particularly energetic decays lead this parent isotope to be the dominant contributor to the RU dose field. Given that these daughters are removed from the uranium stream at the time of separation, it is advantageous to enrich RU as quickly as possible to avoid a costly secondary purification step. Indeed, the dose rate from RU immediately following its separation is very nearly the same as that of NU. One year after separation, the RU dose rate increases to almost ten times that of NU and its decay power exhibits an even more substantial increase; the radiation field from RU peaks about 10 years after separation. Therefore, it seems essential to enrich the RU within a few months of its separation.

If quick re-enrichment is not possible, or if the original separation process does not sufficiently extract certain fission products and actinides, additional “polishing” of the RU would be required. A number of polishing processes have been proposed. While PUREX or a similar aqueous process could be employed, given the low contaminant concentrations other methods offering considerably less complexity and expense can be pursued. One of these is fluoride volatility purification; high-purity
separation of uranium fluorides from fluorides of many fission products and actinides. Uranium fluorides become volatile at significantly lower temperatures than other fluoride compounds; none of the noble metal fluorides become gaseous at a temperature within 30 K of the UF₄ boiling point. Indeed, this purification process is already employed at the Metropolis Works and other U fluorination facilities, and the cost of purifying RU in this fashion would be similar to the cost of conversion.

To ensure that the tails from the RU enrichment process can be stored and disposed in the same manner as DU, it is advantageous to consider three additional options for RU, in addition to those given above for DU.

F. Blending of RU feedstock with NU to decrease ²³²U and ²³⁵U concentrations.
   i) this strategy offers the further advantage of reducing the level of over-enrichment required to compensate for the negative effect of U-236 on the neutron economy of a reactor.

G. Employment of a secondary cascade to produce a second tails stream that is highly concentrated in ²³⁵U.  
   i) this small amount of material would require some decades of storage before becoming disposable in the same fashion as traditional DU

II. Over-enrichment of NU followed by direct blending with RU 
   i) Under this option, RU would not pass through an enrichment plant at all
   ii) Benefits – marginal reductions in NU and SWU requirements per unit energy produced – are minimal

This paper documents DU and RU inventories and lays out the space of options connected to the disposal or re-use of these materials. Future work will include levelized cost-benefit analyses of the options. These analyses will seek to identify the NU and SWU prices at which the various options become competitive with a baseline strategy of immobilization followed by disposal. It is expected that some of the re-use options may serve as valuable hedges against periods of elevated primary uranium prices.

ACKNOWLEDGMENTS

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REFERENCES


2. Cost estimate based on communication with Uranium Disposition Services, LLC.


