Y/ES-261

LOCKHEED MARTIN

Characterization Summary of Candidate Off-Specification Material for Transfer to the Tennessee Valley Authority

Highly Enriched Uranium
Disposition Program Office

January 1998

Prepared by the Oak Ridge Y-12 Plant
Oak Ridge, Tennessee 37831, managed by
Lockheed Martin Energy Systems, Inc.,
for the U.S. DEPARTMENT OF ENERGY
Under contract DE-AC05-84OR21400

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1.0 BACKGROUND

The United States Enrichment Corporation (USEC) Privatization Act, Public Law 104-134, Sec. 3112(e)(1), signed April 26, 1996, provides that, prior to privatization, the Department of Energy (DOE) may transfer off-specification (off-spec) enriche d uranium to a federal agency if the material is transferred for the use of the receiving agency without resale or subsequent transfer to another entity. The Tennessee Valley Authority (TVA) has expressed an interest in obtaining some of this off-spec surplus highly enriched uranium (HEU) for the purpose of making fuel for its reactors. Discussions between DOE and TVA over the past two years have resulted in a framework between DOE and TVA within which to develop a program for the utilization of HEU blended to off-spec low-enriched uranium (LEU) for use in TVA commercial nuclear power reactors. A minimum of 30 metric tons uranium (t) of HEU has been designated for this program.

DOE has identified material amounting to approximately 35 t that potentially will be transferred to the TVA. This material is located at several domestic HEU processing and storage facilities, both within the DOE and in the private sector.

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2.0 TYPE I MATERIAL: OFF-SPEC HEU URANYL NITRATE FROM REPROCESSED IRRADIATED FUEL (9.1 t)

The Type I material consists of 9.1 t of surplus off-spec HEU, comprised of 1.3 t contained in dilute reprocessing solutions of uranyl nitrate and 7.8 t contained in aluminum-clad alloy (UAl_x) irradiated reactor fuel. Because of its processing history, all of this material has similar concentrations of uranium isotopes and should have similar elemental concentrations (after reprocessing of the irradiated HEU reactor fuel).

2.1 Material Characteristics

HEU Solutions (1.3 t): This inventory consists of 230,000 liters of dilute uranyl nitrate solutions containing 1.3 t of HEU that remained after reactor fuel reprocessing operations were suspended. The average assay of the HEU in these solutions is 61 percent ^{235}U. The uranium isotopic analysis of these HEU solutions is shown in Table 1. The concentration of both ^{234}U and ^{236}U will exceed ASTM specification limits after blending.

Table 1. Uranium Isotopic Analysis of HEU Solutions

Uranium Isotope	% of Total U[1]	Concentration[1]
^{232}U	0.000002	3.3×10^{-2} µg/g ^{235}U
^{233}U	0.01	165 µg/g ^{235}U
^{234}U	1.2	20,400 µg/g ^{235}U
^{235}U	60.5	N/A
^{236}U	23.7	392,000µg/g ^{235}U

[1]Obtained from NMC&A test sample taken in December 1995.

A chemical analysis of the HEU solutions is shown in Table 2. Because of the processing history, these materials likely contain small amounts of neptunium and plutonium. However, equipment used in the analysis was not adequately sensitive to determine the neptunium and plutonium content. The proposed alpha activity limit for plutonium and neptunium for off-spec LEU is under development. It is likely that the acceptable upper limit will be less than 50 Bq/gU. However, this material appears to be suitable as off-spec fuel.

Table 2. Chemical Analysis of HEU Solutions

Chemical Element	Concentration[1] (μg/gU)
Aluminum	<0.4
Calcium - Magnesium	10.8
Chromium	<0.4
Cobalt	<1.0
Iron	3.6
Nickel	<0.4
Plutonium - Neptunium	>0 [2]
Silicon	<2.0
Thorium	<3.0

[1]Obtained from NMC&A test sample taken in December 1995.
[2]This material is presumed to contain plutonium and neptunium. Equipment used in this analysis was not adequately sensitive to determine plutonium content.

HEU Reactor fuel (7.8 t): This inventory consists of approximately 3,000 irradiated fuel elements. The fuel in these elements is an Al-clad, UAlx alloy. Historically, much of this fuel has been reprocessed many times. No laboratory analyses are available for this irradiated material. However, Table 3 shows the uranium isotopics from 1,896 irradiated fuel elements that have been calculated from reactor batching memoranda that give the isotopic composition of fuel charged to reactors and the fuel exposure at discharge.

Table 3. Data for Irradiated HEU Reactor Fuel

Batch	A	B	C	E	F	G
No. of cycles in reactor	5	2	4	3	2	1
Days fuel cooled on 01/01/95	2835	2328	2735	2383	2459	884
Assemblies[1]	96	432	252	252	432	432
U Isotope, %						
U-234	1.3	1.3	1.3	1.3	1.3	1.3
U-235	54.8	58.8	59.2	60.0	64.2	65.8
U-236	28.0	18.3	24.4	23.8	21.5	19.7
U-238	15.9	21.6	15.1	14.9	13.0	13.2

[1]Information on a total of 1896 fuel elements (assemblies) is presented in this 1995 table. Current records list 1883 fuel elements available.

Isotopic and trace element data from the 1.3 t in uranyl nitrate solution form (see Tables 1 and 2) is used to represent the expected chemical composition of the 7.8 t of reactor fuel after it has been processed. This assumption is reasonable since the 7.8 t of reactor fuel will be processed similar to the manner in which 1.3 t in solution form was processed. The HEU solutions (previously described) are derived from chemically processing Al-clad, UAl_x alloy reactor fuel. Therefore, the chemical composition of the HEU derived from this reactor fuel, when chemically processed, is expected to be similar.

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3.0 TYPE II MATERIAL: OFF-SPEC HEU URANIUM–ALUMINUM ALLOY (14.6 t)

The Type II material consists of 14.6 t of UAl_x alloy. The UAl_x is material that has been irradiated, reprocessed, blended with higher assay HEU to increase enrichment, and fabricated into UAl_x alloy (nominally 25 percent uranium and 75 percent aluminum) for subsequent fabrication of new fuel elements. This material has not been irradiated since it was last reprocessed.

3.1 Material Characteristics

UAl_x alloy: The UAl_x material consists of a combination of UAl_x ingots and fresh reactor fuel elements. There are approximately 7,900 ingots and 3,100 reactor fuel elements. The ingots were produced by melting and casting a variety of the types of the reactor fuel elements. The net weight of ingots averages about 5 kgs each and the net weight of the larger-sized fuel elements averages around 12 kgs each. The larger fuel elements consist of approximately 44 percent 8001 Aluminum, 33 percent pure (99.99 percent) Aluminum, 7 percent 5052 Aluminum, and 16 percent HEU metal. Table 4 shows approximate maximum impurities for the larger fuel elements and the UAl_x ingots. These impurity values are calculated from process specifications and may not be fully representative of all of the UAl_x material. The information is provided for reference only.

Table 4. Maximum Calculated Chemical Impurities for UAl_x

	Max. Impurities per Item (gms)	
Contaminant	Larger Fuel Element	Ingot
Antimony	0.08	0.06
Beryllium	0.06	0.06
Boron	0.12	0.10
Cadmium	0.33	0.27
Calcium	0.20	0.19
Carbon	1.20	1.12
Chromium	2.80	0.68
Cobalt	0.12	0.10
Copper	9.10	8.94
Iron	42.00	40.21
Lithium	0.84	0.70
Magnesium	40.40	5.81
Manganese	1.00	1.57
Nickel	71.00	64.84
Silicon	12.38	11.54
Sodium	0.05	0.05
Zinc	3.50	4.17
Others	9.30	8.42
Total	194.48	148.83

Note: The weight of each constituent was multiplied by the maximum acceptable weight percent of various items permitted in the specification.

As with the other off-spec HEU, the UAl_x material requires conversion into a readily usable form for LEU commercial reactor fuel. The exact processing methodology will depend upon the blending process utilized. In the past, HEU has been recovered from this type of alloy by first dissolving the alloy in NaOH solution, filtering the solution to capture the insoluble uranium compound, dissolving the solids in nitric acid, purifying the resultant uranyl nitrate solution by solvent extraction, evaporating the purified solution, denitrating to UO_3, reducing the UO_3 with hydrogen to make UO_2, hydrofluorinating to UF_4, and reducing the UF_4 with calcium to make pure uranium buttons.

A portion of the ingot material has been analyzed for purity and isotopic abundance. Averages of the analyses of 2.6 t of UAl_x alloy ingots are provided in Table 5.

The uranium isotopic concentrations vary slightly between individual batches, with average concentrations at 19.9 percent ^{236}U, 1.2 percent ^{234}U, 66 percent ^{235}U, and 13.4 percent ^{238}U. Chemical purity between individual batches vary by a factor of two. It is therefore assumed that the aluminum content of the alloy ranges from 70 to 85 percent. The concentration of ^{232}U and transuranics in this material should be similar to uranium metal buttons (refer to the Type III material section).

Table 5. Analyses of UAl_x Ingots

Avg. Ingot Net Wgt (g)	No. of Ingots	Avg. Purity (gU/g)	Avg. U Isotope Abundances (wt %)		
			^{234}U	^{235}U	^{236}U
5458	96	.2935	1.18	65.82	19.81
5503	96	.3047	1.18	65.97	19.71
5305	96	.2927	1.19	69.70	19.91
5428	96	.2978	1.19	66.06	19.67
5564	96	.2996	1.20	65.27	20.25
5485	96	.3013	1.19	65.84	19.84
5467	96	.2922	1.19	65.77	19.84
5245	96	.3022	1.19	65.21	20.19
5663	96	.3193	1.21	64.18	21.08
5438	96	.2899	1.18	65.89	19.75
5256	96	.3169	1.18	65.94	19.75
5076	96	.2652	1.21	63.11	21.79
4795	96	.1549	1.17	65.48	19.69
4672	96	.1608	1.17	65.96	19.41
4681	96	.1530	1.17	65.28	19.79
4962	96	.1600	1.17	65.04	20.00
5132	96	.1715	1.17	64.59	20.25
5171	96	.1713	1.15	66.03	19.20
5046	96	.1621	1.17	64.91	19.98
5068	96	.1676	1.16	64.41	18.62
5073	96	.1714	1.18	65.34	19.70

4.0 TYPE III MATERIAL: OFF-SPEC HEU UNALLOYED METAL (9.6 t)

The Type III material consists of 9.6 t of HEU in the form of pure metal buttons. These metal buttons were the feed stream to the same HEU fuel fabrication process that produced the UAl_x alloy described in the previous section. On average, these buttons contain about 4.5 kgU each.

4.1 Material Characteristics

The uranium isotopic concentrations vary slightly between individual buttons, with average concentrations at 1.3 percent ^{234}U, 55.0 percent ^{235}U, and 27.6 percent ^{236}U. It is assumed that the remainder is ^{238}U. Recent health physics surveys on 30 storage containers (each containing four buttons) measured surface activity on the containers ranging up to 100 mr/hr.[i] Measured radiation levels 30 cm away from containers range between 15 and 20 mr/hr.

Approximately 560 samples of this material have been analyzed for isotopic abundance and chemical impurities. The results, given in Table 6, reflect the actual characteristics of 88 percent of the 9.6 t inventory of this material.

Table 6. Analyses of Unalloyed Metal Buttons

Element	Units	Mean	Minimum	Maximum
Uranium	wt %	99.9409	99.3450	99.9562
U-232	µg/gU	0.0202	0.0113	0.0287
U-234	wt. %	1.28	1.2	1.5
U-235	wt. %	55.46	45.19	64.87
U-236	wt. %	28.13	20.41	36.41
U-238	wt. %	16.27	13.46	20.16
Np/Pu	Bq/gU	198.15	20.33	1,003.33
Al	ppm	20.6	1	120
As	ppm	<10	*	*
Au	ppm	<1	*	*
B	ppm	0.47	0.1	5.6
Ba	ppm	2.01	0.1	12
Be	ppm	0.11	0.1	2
Bi	ppm	1.02	1	4
C	ppm	117	35	315
Ca	ppm	17.6	10	406
Cd	ppm	0.121	0.1	0.5
Co	ppm	1.10	1	5
Cr	ppm	9.40	0.1	41
Cu	ppm	7.6	1	184
Fe	ppm	40.8	11	345
Ga	ppm	<1	*	*
Ge	ppm	<1	*	*
Li	ppm	0.37	0.1	4.5
Mg	ppm	16.5	1	87
Mn	ppm	5.39	1	22
Mo	ppm	<10	*	*
Na	ppm	1.08	1	40
Nb	ppm	14.6	10	51
Ni	ppm	16.5	1	146
P	ppm	<100	*	*
Pb	ppm	4.46	4	10
Pd	ppm	<1	*	*
S	ppm	<2	*	*
Sb	ppm	<2	*	*
Si	ppm	22.8	10	347
Sn	ppm	10.02	10	19
Sr	ppm	<20	*	*
Ti	ppm	4.5	4	34
V	ppm	1.35	.07	9
W	ppm	<100	*	*
Zn	ppm	<10	*	*

*Indicates that values are equal to the mean item value.

Data suggest that these metal buttons will not require additional purification before blending. A few samples show high levels of neptunium and plutonium, but the mean value for these contaminants is well below the proposed limit. Individual lots with high levels of plutonium and neptunium might be handled by blending with other lots having much lower levels. Total gamma activity was not reported, but the ^{232}U concentrations were about 1 percent of the level that would be required for decay of ^{232}U and its daughters to exceed the ASTM specification limit on gamma from reprocessed uranium. The non-radioactive elements present were all at concentrations below the proposed specification.

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5.0 TYPE IV MATERIAL: OFF-SPEC HEU MISCELLANEOUS MATERIALS (1.4 t)

The Type IV material consists 1,378 kgU of miscellaneous materials in forms such as floor sweepings and process residues. The materials in the miscellaneous category have a similar processing history to the Type II material. This material is included because it completes the inventory of this same processing history material. However, some of this material is not immediately available and will require processing prior to packaging and shipment.

5.1 Material Characteristics

Table 7 presents a summary of the miscellaneous materials by form.

Table 7. Off-Spec HEU Miscellaneous by Form

Materials	Total (kgU)
Misc. - Solids	551.8
Metal - Alloyed Scrap	404.4
Oxide - Other	165.3
Oxide - UO_3	152.1
Solutions - Aqueous	36.6
Compounds - UF_4	35.5
Sources & Standards - Standards	12.4
Solutions - Organic	11.4
Oxide - U_3O_8	3.9
Misc. Equip Holdup	2.6
Sources & Standards - Samples	0.6
Misc. - Discard	0.3
Compounds - UNH	0.3
Misc. - Other	0.2
Combustibles - Other	0.2
Total	1,377.6

Note: Totals may not sum due to rounding.

The 404.4 kgU of alloyed metal scrap is the inventory of surplus HEU originally reserved for TVA's lead use assemblies (LUA) for the demonstration. The uranium isotopic concentrations in this material is 66 percent ^{235}U, 19 percent ^{236}U, 1.2 percent ^{234}U, 10 ppm ^{233}U, and 20 ppb ^{232}U. The remainder, approximately 14 percent, is ^{238}U.

Three samples of this 404.4 kgU inventory have been purified (through a laboratory-scale solvent extraction process), combined, and analyzed. The results are provided in Table 8. Because of sampling techniques, the solvent extraction process utilized, and sensitivity of the analytical equipment, this sample may not accurately represent LEU derived from this miscellaneous HEU. Therefore, Table 8 is included for information purposes.

Excepting the 404.4 kgU of material in the TVA LUAs, other miscellaneous materials have not been adequately characterized. These materials exist in a variety of forms including impure oxides, crucible "skull" oxide, trichlorethane oil, ceramic crucibles, concentrated nitrate solutions, dilute nitrate solutions, carbitol and tri-butyl phosphate, tetrafluoride, clinkers and screenings, ash and other unleached solids, and discard solids. The ^{235}U assay of these materials ranges from 50 to 85 percent. Other uranium isotope concentrations vary widely. In general, these miscellaneous HEU materials will need to be purified and converted to suitable forms such as oxide compounds or metal.

Table 8. Analysis of LUA Material

Element	Unit	Results
^{238}Pu	pCi/g	<12,900
^{239}Pu	pCi/g	<12,900
^{232}U	pCi/g	300,000
^{234}U	wt %.	1.18
^{235}U	wt %	63.78
^{236}U	wt. %	20.20
^{238}U	wt. %	14.83
^{99}Tc	$\mu g/g\ ^{235}U$	0.06
Al	ppm U basis	>500
Sb	ppm U basis	<3
As	ppm U basis	<5
B	ppm U basis	0.68
Br	ppm U basis	<2.5
Cl	ppm U basis	20
Cr	ppm U basis	<3
Gd	ppm U basis	<0.2
Sm	ppm U basis	<0.2
Dy	ppm U basis	<0.2
Eu	ppm U basis	<0.2
Mo	ppm U basis	<2.0
Nb	ppm U basis	n/a
P	ppm U basis	230
Ru	ppm U basis	n/a
Ta	ppm U basis	n/a
Ti	ppm U basis	<50
W	ppm U basis	n/a
V	ppm U basis	<5
Zr	ppm U basis	n/a
Hg	ppm total weight basis	8.5

(1) The data for the enrichment and mercury is a weighted calculation from the results of the three feed samples. All other values were obtained on a sample of oxide generated from a composite of the three feed samples.

(2) The ^{232}U, ^{99}Tc, Br, and Cl numbers are on a best-effort basis. These analyses were done without an actual procedure but were analyzed by supervisors using established protocol.

(3) The high Al and P numbers are most likely due to carryover from the solvent extraction process in the lab and do not represent levels expected in a uranium recovery facility.

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ABBREVIATIONS, ACRONYMS, AND INITIALISMS

ASTM	American Society for Testing and Materials
CADS	Characterization Analysis Database System
DOE	Department of Energy
HEU	highly enriched uranium
LEU	low-enriched uranium
LUA	lead use assembly
MOU	memorandum of understanding
NMC&A	nuclear materials control and accountability
NRC	Nuclear Regulatory Commission
TVA	Tennessee Valley Authority
UAl_x	Uranium-Aluminum Alloy
USEC	United States Enrichment Corporation

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CHEMICAL NOTATIONS

Al	aluminum	As	arsenic
Au	gold	B	boron
Ba	barium	Be	beryllium
Bi	bismuth	Br	bromine
C	carbon	Ca	calcium
Cd	cadmium	Cl	chlorine
^{36}Cl	chlorine isotope 36	Co	cobalt
Cr	chromium	Cs	cesium
^{135}Cs	cesium isotope 135	^{137}Cs	cesium isotope 137
Cu	copper	Dy	dysprosium
Eu	europium	Fe	iron
Ga	gallium	Gd	gadolinium
Ge	germanium	Hg	mercury
Li	lithium	Mg	magnesium
MgF_2	magnesium fluoride	Mn	manganese
Mo	molybdenum	Na	sodium
NaF	sodium fluoride	NaOH	sodium hydroxide
Nb	niobium	Ni	nickel
^{59}Ni	nickel isotope 59	^{63}Ni	nickel isotope 63
Np	neptunium	^{237}Np	neptunium isotope 237
P	phosphorus	^{231}Pa	protactinium isotope 231
Pb	lead	Pd	palladium
Pu	plutonium	^{238}Pu	plutonium isotope 238
^{239}Pu	plutonium isotope 239	PuO_2	plutonium dioxide
Ru	ruthenium	S	sulfur
Sb	antimony	Se	selenium
Si	silicon	Sm	samarium
Sn	tin	Sr	strontium
^{89}Sr	strontium isotope 89	^{90}Sr	strontium isotope 90
Ta	tantalum	Tc	technetium
^{99}Tc	technetium isotope 99	Ti	titanium
U_3O_8	triuranium octaoxide	UAl_x	uranium-aluminum alloy
UNH	uranyl nitrate hexahydrate	UF_4	uranium tetrafluoride
UF_6	uranium hexafluoride	UO_2	uranium dioxide
$UO_2(NO_3)_2$	uranyl nitrate	UO_3	uranium trioxide
^{232}U	uranium isotope 232	^{233}U	uranium isotope 233
^{234}U	uranium isotope 234	^{235}U	uranium isotope 235
^{236}U	uranium isotope 236	^{238}U	uranium isotope 238
V	vanadium	W	tungsten
Zn	zinc	Zr	zirconium

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UNITS OF MEASURE

Bq/gU	becquerels per gram of uranium
cm	centimeter
dpm/g	disintegrations per minute per gram of material
gms	grams
kgU	kilogram of uranium
µg	microgram
mg/g	milligrams per gram of material
mr/hr	millirems per hour
pCi/g	picocuries per gram
ppb	parts per billion on a weight basis
ppm	parts per million on a weight basis
t	metric ton of uranium
%	weight-% (when used to specify an isotopic assay)

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