AIIM

Association for Information and Image Management

1100 Wayne Avenue, Suite 1100
Silver Spring, Maryland 20910
301/587-8202

Centimeter

Inches

MANUFACTURED TO AIIM STANDARDS
BY APPLIED IMAGE, INC.

O

Document No. SAC200038940000

DOCUMENT NO.

HW-67475-DEL

Declassified With Deletions

This document consists of 1011
(CLASSIFICATION) Number 1 of 1 Copies, Series SA 20A

GENERAL ELECTRIC

HANFORD ATOMIC PRODUCTS OPERATION - RICHLAND, WASHINGTON

DATE

November 21, 1960

TITLE

SUPPLEMENTARY BIRCH PRODUCTION (u)

OTHER OFFICIAL CLASSIFIED INFORMATION

THIS MATERIAL CONTAINS INFORMATION AFFECTING THE NATIONAL DEFENSE OF THE UNITED STATES WITHIN THE MEANING OF THE ESPIONAGE LAWS, TITLE 18, U. S. C., SECS. 793 AND 794, THE TRANSMISSION OR REVELATION OF WHICH IN ANY MANNER TO AN UNAUTHORIZED PERSON IS PROHIBITED BY LAW.

AUTHOR

L. W. Lang &
B. F. Judson

THIS DOCUMENT MUST NOT BE LEFT UNATTENDED OR WHERE AN UNAUTHORIZED PERSON MAY HAVE ACCESS TO IT. WHEN NOT IN USE, IT MUST BE STORED IN AN APPROVED LOCKED REPOSITORY WITHIN AN APPROVED GUARDED AREA. WHILE IT IS YOUR POSSESSION AND UNTIL YOU HAVE OBTAINED A SIGNED RECEIPT FROM CLASSIFIED FILES, IT IS YOUR RESPONSIBILITY TO KEEP IT AND ITS CONTENTS WITHIN THE LIMITS OF THIS PROJECT AND FROM ANY UNAUTHORIZED PERSON. ITS TRANSMITTAL TO, AND STORAGE AT YOUR PLACE OF RESIDENCE IS PROHIBITED. IT IS NOT TO BE DUPLICATED. IF ADDITIONAL COPIES ARE REQUIRED, OBTAIN THEM FROM THE RELATED ISSUING FILE. ALL PERSONS READING THIS DOCUMENT ARE REQUESTED TO SIGN IN THE SPACE PROVIDED BELOW.

ROUTE TO:	PAYROLL NO.	LOCATION	FILES ROUTE DATE	SIGNATURE AND DATE
L. W. Lang	12926	170417	11-23-60	L. W. Lang 4-23-61
L. W. Lang	12926	Rec. 82616	11-23-60	L. W. Lang 4-24-61
L. W. Lang	12926	170417	11-23-60	L. W. Lang 4-24-61
R. L. Stutheit	OCT 22 1992			
AA Lorisgalli	SA-232			
[REDACTED]				
[REDACTED]				
R. L. Stutheit	MAR 4 1994			
[REDACTED]				
[REDACTED]				

CIRCULATING COPY

OCT 22 1992

CIRCULATING COPY

MAR 4 1994

RECEIVED-DOE-RI
DOCUMENT CONTROL

Declassified With Deletions

Declassified With Deletions

This document consists of 10 pages.

SUPPLEMENTARY BIRCH PRODUCTION

By

L. W. Lang
and
B. F. Judson

November 21, 1960

Classification cancelled (Change to

UNCLASSIFIED

By Authority of CG-PR-2

C. R. Bauman, PNL
Classification Officer
By 3-4-94

Verified By A. L. Solta 3-22-94
J. K. Ruel 3-22-94

DISTRIBUTION

1-3. HOO-AEC: Attn.: A. T. Gifford	14. W. K. MacCready
4. J. H. Brown	15. L. H. McEwen
5. H. A. Carlberg	16. W. N. Mobley
6. V. R. Cooper	17. H. M. Parker
7. R. L. Dickeman	18. C. A. Priode
8. O. H. Greager	19. O. C. Schroeder
9. A. B. Greninger	20. H. P. Shaw
10. C. T. Groswith	21. J. H. Warren
11. W. E. Johnson	22. H. F. Zuhr
12. B. F. Judson	23. 300 Files
13. L. W. Lang	24. Records Center

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

ds
DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Declassified With Deletions

(att 1)

SUPPLEMENTARY BIRCH PRODUCTIONINTRODUCTION

In response to specific requests of the AEC and as part of a Combined Operations over-all review, a number of engineering studies have been made of alternative methods for increasing availability of neptunium at Hanford. Some of the alternatives that have been considered include special processing of neptunium mined from existing stored wastes, improving recovery methods from separations plants, maximizing inventory usage by close-coupled separations-reactor systems, and recycling U^{236} contained in the natural and enriched uranium streams discharged from the Hanford reactors^(1,2,3,4,5). The goal of the studies has been to provide substantial amounts of neptunium in addition to that forecasted for the Hanford and Savannah River sites to meet the inventory needs of the AEC over the next four to eight years. Conditions important to the studies have been the timeliness, cost, practicality, and program development needs in providing the increased neptunium. The following report presents results of recent studies on one of the more promising of these schemes - recycling E-metal at Hanford.

The report updates an earlier study in which recycling of both natural and enriched uranium was considered for Hanford⁽⁵⁾. The earlier study showed that recycled natural uranium would provide appreciable gains in neptunium availability but at an excessive cost. Recycle of the slightly enriched uranium streams proved a more economical means of realizing smaller but still significant gains in neptunium production. Subsequent to the earlier report, a feasible and immediately applicable scheme for UF_6 blending has been conceived demonstrating further advantages for recycling the enriched uranium.

SUMMARY, CONCLUSIONS AND RECOMMENDATIONS

Approximately 34 kilograms of supplementary neptunium could be produced at Hanford during the next seven to eight years by upgrading irradiated E-metal and NPR uranium through a blending operation at Oak Ridge (rather than in the diffusion cascades) and then recycling the material through the Hanford reactors. (cf Table 1) Such a scheme would conserve uranium-236 for use as a source of neptunium-237 in the reactors without incurring major capital costs. Oak Ridge Operations personnel have estimated that capital needs for the requisite UF_6 blending operation would not exceed \$50,000 (Ref. 6). No major costs are foreseen for other sites within the AEC complex, including Hanford and Fernald. In addition, the program would not require extensive development efforts to demonstrate technical feasibility and would not interfere appreciably with normal fuel cycle operations. Production-scale recycling of E-metal could thus start immediately.

TABLE NO. 1

SUPPLEMENTARY BIRCH PRODUCTION

- Case 1: Blended material charging starts January 1, 1961.
 2: Blended material charging starts January 1, 1962.

kg Supplemental Birch Separated*
From NPR and Old Reactor Slightly Enriched U

	<u>Case 1</u>	<u>Case 2</u>
FY-63 (Cumulative)	1.78	.46
FY-64 (Cumulative)	5.34	1.90
FY-65 (Cumulative)	10.81	5.73
FY-66 (Cumulative)	17.56	11.12
FY-67 (Cumulative)	25.26	17.76
FY-68 (Cumulative)	33.76	25.40

*Assumes Redox Recovery of 0.9.

COSTS

	<u>Total</u>	<u>Unit</u>	<u>Total</u>	<u>Unit</u>
Capital	50,000		50,000	
kg of Top-product @ 12	6,045,000	179.00	5,094,000	200.50
Blending Costs @ .005/#	110,000	3.30	89,000	3.50

DOE b(3)

DOE b(2)

Declassified With Deletions

Declassified With Deletions

Incremental costs anticipated for the additional 34 kilograms of neptunium are approximately \$3 per gram of neptunium for off-site out-of-pocket costs. In addition, there would be a reduction in the amount of highly enriched uranium produced in the diffusion cascades of approximately 15 grams of uranium-235 per gram of neptunium. There would also be a reduction in the plutonium output from the Hanford reactors conservatively estimated at 0.9 grams of plutonium per gram of additional neptunium. The amount of neptunium gained through a recycle program could be further increased and the over-all unit costs decreased by future revisions in reactor usage patterns for slightly enriched uranium and through optimization of the blending operation that would minimize uranium-236 losses. The program is flexible enough to permit realizing or foregoing such gains, depending upon forecasted needs. Realization of the additional 34 kilograms of neptunium by the end of FY-1968, however, would be dependent upon starting the program as soon as practical to permit recycling an existing E-metal inventory and accumulating a suitable inventory for the NPR.

The study has shown that an E-metal recycle program at Hanford is a highly attractive method for increasing neptunium availability at reasonable cost. The program would have high assurance of success in supplementing long-range neptunium needs and at the same time would permit flexibility in the development of other product programs. It is therefore recommended that an E-metal recycle program be started at Hanford as soon as practical.

Study Bases

Neptunium-237 is currently produced in the Hanford reactors by two mechanisms: (1) the $n, 2n$ reaction of U^{238} to form U^{237} , which beta decays to neptunium-237, and (2) the successive n, γ reactions with U^{235} to form U^{236} and then U^{237} , which beta decays to neptunium-237. Cross section values and relative yields from the reactions are discussed in more detail in HW-65072(5). The available estimates indicate that in the exposure range of interest, 75 to 80 percent of the neptunium formed in the reactors results from the $n, 2n$ reaction. An E-metal recycle program would increase the proportion of neptunium produced by the n, γ reactions by conserving the U^{236} normally produced in the reactors to permit further neutron capture, forming U^{237} and thus neptunium-237.

The fundamental recycle schemes evaluated as means of increasing neptunium availability consist of:

1. Processing irradiated E-metal and NPR uranium through the Redox Separations Plant at Hanford for routine recovery of neptunium, plutonium, and uranium.

2. Upgrading the uranium to the 0.947 percent U^{235} concentration required for reactor charging by blending the irradiated uranium as UF_6 with 1.2 or 1.4 percent U^{235} at Oak Ridge, thereby bypassing the diffusion cascades and conserving the U^{236} .
3. Processing the recycled uranium through the normal reduction and fabrication operations at Fernald, and the canning operation at Hanford.
4. Charging the recycled uranium into the reactors in the normal usage pattern for slightly enriched uranium.
5. Discharging, cooling and processing the recycled uranium in the normal manner to start another cycle.

Two cases were studied to evaluate the effects of timing in starting a recycle program. In case one, recycling uranium was assumed to start in January 1961 and in case two, in January 1962. A twelve-month fuel cycle was assumed, approximating present cycling timing: four months for irradiation and three months cooling time, plus one month for each processing step for separation, blending, fabrication, canning and pile charging. Production calculations were based on E-metal throughputs for existing reactors of 960 tons per year through FY 1964 and 1010 tons per year thereafter to FY 1968. First blending for NPR was assumed to occur in the fourth quarter of FY 1963, with a throughput rate of 800 tons per year. Exposure levels were assumed to be 800 MWD/T for E-metal and 1200 MWD/T for NPR. These assumptions approximate current "base-case" production forecasts.

U^{236} and Np^{237} buildup calculations were based on the approximate methods given by Arnold in ORNL-2104, with certain revisions in assumed cross section values⁽⁷⁾. As discussed in HW-65072, effective thermal cross sections more appropriate to the existing Hanford reactors are 8.1 millibarns for the U^{238} n, 2n reaction, 37.2 ± 8 barns for the U^{236} n, γ reaction, and 220 ± 20 barns for the Np^{237} burnout reaction⁽⁵⁾. The respective thermal cross section values for the NPR are estimated to be 16.2 ± 3 millibarns, 73 ± 17 barns, and 220 ± 20 barns. Normal production of E-metal at an exposure of 800 MWD/T would thus be 2.0 grams per ton, of which 1.5 grams per ton results from the n, 2n reaction, and 0.5 grams per ton from the n, γ reactions. Similarly, for NPR at 1200 MWD/T exposure, assuming neptunium concentration is 5.2 grams per ton with 3.6 grams per ton due to the n, 2n reaction, and 1.6 grams per ton due to the n, γ reaction. U^{236} concentrations for E-metal and NPR are estimated to be 130 and 189 grams per ton, respectively. The effects of U^{236} recycle and buildup were approximated by assuming straight line extrapolations, correcting for blending discards but neglecting U^{236} burnout. Although neglecting U^{236} burnout slightly over-states the production gains, the extrapolation method is conservative and actual gains may be larger than indicated.

Neptunium Gains

The estimated increase in neptunium availability resulting from the two U²³⁶ recycle cases are summarized in Table 1. Approximately 34 kilograms of supplementary neptunium could be produced at Hanford during the next seven to eight years if the E-metal recycle program were to start immediately. Starting immediately would permit recycling an existing inventory of irradiated E-metal (approximately 400 tons). This recycle material will be adequate in quantity to fill the fuel cycle pipeline to meet E-metal requirements for existing reactors as well as charge the NPR in FY-1964.

The neptunium gains presented in Table 1 are larger than those previously estimated in HW-65072, principally due to earlier charging of NPR with recycled material and the use of both 1.2 and 1.4 percent U²³⁵ to upgrade the depleted material. The U²³⁶ buildup and thus the neptunium-237 production level is dependent upon the concentration of U²³⁵ used to upgrade irradiated uranium back to 0.947 percent U²³⁵. Use of 1.2 percent U²³⁵ involves lower degradation charges initially but for a constant E-metal load, discards a greater fraction of irradiated uranium in the blending operation and thus reaches the lower equilibrium level of U²³⁶ more rapidly. As a result, neptunium production gains are lower and average unit costs higher. Using 1.4 percent U²³⁵ material, however, increases the immediate degradation charges but results in higher equilibrium levels of U²³⁶ and higher neptunium gains at a lower average unit cost. Both case studies were therefore based on use of 1.2 percent U²³⁵ until recycle of the NPR was started and then using 1.4 percent U²³⁵ for the remainder of the study period. Use of 1.2 percent U²³⁵ is preferred prior to NPR start-up since E-metal needs will be expanding and there would be no gain in employing a higher enrichment.

Further increases in neptunium production could be realized with the simultaneous reduction in over-all unit costs if the reactor usage pattern for slightly enriched uranium were revised, and if the blending operation were optimized. Thus blanket loading and overboring programs in the existing reactors could gradually increase the E-metal usage such that no U²³⁶ would be lost in discards from the blending operation during the next few years and therefore the U²³⁶ inventory would build up to a higher equilibrium level with resultant gains in neptunium production. Furthermore, the concentration of U²³⁵ employed in the blending operation could be increased to some optimum value above 1.4 percent, further conserving U²³⁶ and realizing neptunium production gains. In addition, U²³⁶ contained in the discards from the blending operation could be irradiated on a "once-through" basis by blending the material into the natural uranium stream. Although this material would be eventually lost to the diffusion cascades, some burnout to neptunium-237 would be realized. An E-metal recycle program would be

flexible enough to permit revisions such as these to the blending operation and consequent adjustments in neptunium production gains at essentially any time the forecasted needs for neptunium would indicate it to be desirable.

Fuel Cycle Effects

An E-metal recycle program at Hanford would have little effect on the overall fuel cycle operations in the AEC complex, except for the blending operation. Recent studies by C. A. Keller at Oak Ridge indicate that blending the irradiated E-metal and 1.2 or 1.4 percent U²³⁵ streams as uranium hexafluoride could be started immediately⁽⁶⁾. In addition, capital costs requirements would be less than \$50,000, and incremental operating costs would amount to only \$0.005 per pound uranium. Details of the blending operation and estimated costs are quoted from Keller's document in Appendix A.

Adoption of an E-metal recycle program would have no major effect on the performance of the Hanford reactors. Plutonium production was conservatively estimated to be decreased by 0.9 grams of plutonium for every additional gram of neptunium formed due to reduced rate of product burnout and improved thermal neutron utilization. E-metal requirements would be increased slightly to compensate for the decreased reactivity associated with U²³⁶ material. Further study is required to determine possible changes in reactor operating characteristics due to the accumulation of heavy element isotopes and possible variations in U²³⁵ assay. These effects would be less significant for a recycle program, involving only enriched fuels, however, than for a total recycle program. The net effect of increasing U²³⁶ cross sections, would appear to improve thermal neutron utilization and thus over-all conversions.

No major problems are foreseen in the chemical processing of recycled uranium streams. The planned neptunium recovery procedures for the Redox Plant would not be appreciably affected by a two- to three-fold increase in neptunium throughput. Slightly longer cooling times could well be required due to the high U²³⁷ content of discharged metal. No major increase in processing requirements are anticipated to produce a uranium product suitable for refabrication in a recycle program. Preliminary studies indicate that the increased plutonium-238 formed during FY-1963 through FY-1968 in a U²³⁶ recycle program would result in less than 10 percent increase in the neutron and gamma radiation levels of plutonium product. The shipping, reduction, fabrication, handling and charging operations throughout the remainder of the over-all fuel cycle are expected to remain largely unaffected by an E-metal recycle program based on hexafluoride blending.

Neptunium Costs

The incremental costs for producing the 34 kilograms of additional neptunium in case one are approximately \$3.30 per gram of neptunium for off-site out-of-pocket dollar costs. Additional costs to Hanford would be nominal with no major effects anticipated on Hanford's pricing structure. In addition there would be a reduction in the amount of highly enriched uranium produced

in the diffusion cascades of approximately 15 grams of U^{235} per gram of neptunium. There would also be a reduction in plutonium output for the Hanford reactors, conservatively estimated at 0.9 grams of plutonium per gram of additional neptunium. Similarly, for the 25 kilograms of neptunium produced by case two recycling, the average off-site out-of-pocket costs would be \$3.50 per gram of neptunium, the average incremental U^{235} loss 17 grams U^{235} per gram of neptunium, and the incremental plutonium loss 0.9 grams plutonium per gram of neptunium.

The only out-of-pocket dollar costs directly ascribable to an E-metal recycle program result from the hexafluoride blending operations (discussed in Appendix A). A relative value curve supplied by G. A. Garrett was employed in calculating the incremental U^{235} costs, assuming a 0.25 percent zero value tails⁽⁸⁾. Incremental plutonium losses are based on a brief, but conservative review of Hanford reactor physics.

Assuming the reduced U^{235} production is valued at \$12 per gram and the reduced

DOE
6(3)

These nominal "costs" compare favorably with estimated incremental costs for recovering neptunium from existing stored wastes⁽¹⁾. In addition, an E-metal recycle program would have a higher assurance of success within the desired time period than a waste mining program or a program to develop close-coupled reactor-separations systems.

The calculations indicate that E-metal recycling could effectively supplement presently planned programs for meeting neptunium demands. Recycling at Hanford also provides programming flexibility in that over-all production schemes within the AEC complex for other products such as Pu^{239} and tritium can be optimized without undesirable restrictions imposed by neptunium demands. It is therefore recommended that an E-metal recycle program be started at Hanford as soon as practical.

B. F. Judson, Senior Engineer
Research & Engineering Operation
CHEMICAL PROCESSING DEPARTMENT

L. W. Lang, Principal Engineer
Manufacturing Section
IRRADIATION PROCESSING DEPARTMENT

LWL:BFJ:cs

APPENDIX A

Quotations from GEH-25196, "Alternative Methods Of Upgrading 0.865 Material", by C. A. Keller, Oak Ridge Operations Office, dated November 2, 1960:

"In accordance with our telephone conversation of October 25, 1960, we are forwarding preliminary cost estimates for upgrading 0.865% U-235 material to the 0.947% level by three methods. In all methods we have assumed that 1.2% material would be blended with 0.865% material and that the annual requirement for each would be 500 tons U and 1500 tons U, respectively. The three methods considered are as follows:"

* * *

"C. Conversion of 0.865 UO_3 to UF_6 at Oak Ridge, withdrawal of 1.2% UF_6 from the cascade which is blended in the liquid phase with the 0.865 UF_6 ; the 0.947 UF_6 is shipped to FMPC and reduced to UF_4 ."

* * *

"Method C

Method C could be placed in operation immediately. The only cost, in addition to those presently being incurred under the current mode of operation, would be a \$0.005/pound U blending cost. The cost actually incurred would be approximately \$0.025/pound U; however, this cost must be compared to \$0.02/pound U now being incurred at Paducah in achieving the desired precision for 0.947 withdrawals. No new equipment would be needed to put this method into operation, however, cylinder rocking equipment and other miscellaneous items of equipment costing a total of \$50,000, or less, may be desirable if the program is to continue for an extended period of time.

This method would necessitate the conversion of all 0.865 oxide to UF_6 and then the conversion of blended 0.947 UF_6 to UF_4 . The cost of annually converting 1500 tons U as UF_4 to UF_6 would not exceed \$0.20/pound U and would probably average between \$0.15 and \$0.18. The budgeted cost of annually reducing 2000 tons of U as UF_6 to UF_4 would be approximately \$0.08/pound U at FMPC."

REFERENCES

- 1) HW-67313 - "Birch Recovery From Stored Waste," by W. K. MacCready, dated November 3, 1960.
- 2) HW-65200 PT1 - "Purex Palm Extraction and Purification Flowsheet and Technical Criteria for Project CG-821," by R. G. Geier and J. P. Duckworth, dated May 20, 1960.
- 3) HW-65562 - "Requirements for Continuous Palm Recovery at Redox," by R. G. Barnes, dated June 7, 1960.
- 4) HW-59682 - "Advantages of Palmolive Alternates," by E. A. Coppinger and E. T. Merrill, dated March 17, 1959.
- 5) HW-65072 - "Special Production Study," by B. F. Judson and L. W. Lang, dated May 20, 1960.
- 6) GEH-25196 - "Alternative Methods of Upgrading 0.865 Material," by C. A. Keller, dated November 2, 1960.
- 7) ORNL-2104 - "Effect of Recycle of Uranium Through Reactor and Gaseous Diffusion Plant on Build-up of Important Transmutation Products in Irradiated Power Reactor Fuels," by E. D. Arnold, dated September 11, 1958.
- 8) KOA-736 - "Re: R(x) Table," by G. A. Garrett, dated September 2, 1960.

DATE

FILMED

7/25/94

END