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CAPABILTY FOR PROCESSING POWER REACTOR FUELS

H. H. Van Tuyl

March 1976

A Hanford Historical Document Released for Public Availability

Pacific Northwest Laboratories

PNL-8154

Date March 11, 1976

To J. L. McElroy

From H. H. Van Tuyl *Van*

Subject Capability for Processing Power Reactor Fuels

Internal Distribution

L. Bray
LL Burger
RL Dillon
JL Green
ER Irish
JH Jarrett
RE Nightingale
AM Platt
DH Siemens
JL Swanson
HH Van Tuyl - LB
EJ Wheelwright
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INTRODUCTION

PNL is engaged in a major ERDA program on solidification of wastes from processing commercial power reactor fuels. Actual radioactive wastes are needed in this program for laboratory investigations and hot cell demonstration runs on waste solidification. These wastes should be made by solvent extraction for best duplication of the wastes expected from actual reprocessing plants. Facilities at Hanford are available in the 325A hot cells, the 324 hot cells, and the Hot Semiworks for solvent extraction processing of power reactor fuels. Various combinations of these facilities may be used to obtain processing rates ranging from a few hundred grams per day to rates approaching 1 tonne per day. The fuel reprocessing capabilities were reviewed to determine the capacities and costs for various options. In the options involving the 325A hot cells, it was assumed that chopped fuel would be delivered to the system at no charge, and the high level waste would be delivered to 324 without prior concentration.

SUMMARY

The reprocessing of commercial power reactor fuels in existing or modified facilities has been examined for four different reprocessing rates and is summarized below:

2 kg per day

Existing capabilities in the 325A hot cells will permit processing up to 2 kg of fuel per day using an 8-stage centrifugal contactor and discarding the uranium-plutonium stream in the organic phase. The principal problems with this system are feed clarification, equipment reliability, and waste disposal. The cost is anticipated to be about $30,000 per run for runs up to 10 kg each. Higher assurance of continued operation could be obtained by fabricating a spare contactor and replacement parts.

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50 kg per day

An increased capacity of up to about 50 kg per day could be achieved by installation of two inch pulse columns in 325A followed by separating the plutonium from the uranium by anion exchange. The existing dissolver system would be nearly adequate. The plutonium would then be sent to ARHCO for further purification and recovery. The first cycle waste would be collected and shipped to 324 without further concentration. The cost will be about $2,000,000 over a nine month period for equipment installation and checkout. After installation and checkout, operating costs will be about $1,500,000 per year. The annual throughput expected in this system will depend on the operating efficiency. At an operating efficiency of 75% the average throughput would be slightly over one tonne per month. One disadvantage of this method of operation is that it will occupy over half of the 325 hot cells, and could conflict with other work in the cells. The major uncertainties in this processing are possible zirconium fires during shearing, offgas cleanup and waste disposal problems.

200 kg per day

An increase to 200 kg per day could be achieved by installing 4 inch diameter pulse columns in the 324 hot cells. Columns of this size are possible in 324 but not in 325A because of the higher operating space available in 324. The major drawback to this arrangement is that installation of a major dissolution and solvent extraction capability in 324 would severely limit the operation of the waste solidification program. An alternate combination of 324 and 325A would be to provide dissolution in 324 and solvent extraction in 325A. However, this would cause solvent extraction in 325A to be the limiting step of the system. Throughput of about 100 kg per day might be achieved in a 2 inch column system, and perhaps 150 kg/day in 2 1/2 inch columns. Two parallel systems of 2 to 2 1/2 inch columns could achieve 200 kg/day. This alternative would still result in some interference with the waste programs because fuel receipt, shearing, dissolving, and waste solidification would all be done in the same facility.

1000 kg per day

The Hot Semiworks which was established as a pilot plant for the Redox and Purex plants at Hanford is potentially available for use in commercial fuels reprocessing. The capacity could approach one tonne per day. The cost for refurbishing the Hot Semiworks for satisfactory operation has not been evaluated accurately. It will cost at least tens of millions and perhaps a hundred million to prepare the facility for operation. Likewise, operating costs would be high, and the need for such large quantities of waste is not clear.

EXISTING FACILITIES

325A Hot Cells

The three small interconnected hot cells in 325A were designed for laboratory and bench scale studies. The largest cell is 7 feet deep, 15 feet wide, and 15 feet high. The other two cells are 7 feet deep, 6 feet wide, and 15 feet high. Each cell is shielded with four feet of high density concrete and equipped with master slave manipulators and one or more windows for viewing. Remotely operated cranes or hoists are installed in each of the cells. The central cask handling area serves all three cells. Several shielded tanks are located in the basement and adjacent vault area with capacities of up to 18,000 gallons. These tanks can be used for handling process streams and waste solutions.

324 Hot Cells

The 324 Building is shared between PNL and HEDL. The cell space described in this paragraph is that which is under PNL control. It consists of four large engineering cells, a central cask handling area, a wet storage basin for underwater loading of casks and storage of radioactive materials, and two shielded vaults with tanks having capacities up to 5,000 gallons. The cells are shielded with either four feet of high density concrete or 4 1/2 feet of normal concrete. They vary in depth from 9 to 22 feet, in width from 19 to 25 feet, and in height from 15 to 34 feet. Each cell is equipped with two or more windows, television, and two of the cells are also equipped with periscopes. All cells are equipped with manipulators and remotely operated cranes. In addition to the hot cell capabilities, the facility contains a large laboratory for bench to prototype scale engineering studies with nonradioactive materials. This cold area consists of 16 modules each of which is 15 feet by 22 feet in area and 11 1/2 feet high.

Hot Semiworks

The Hot Semiworks facility is located in 200E area and was established as a pilot plant for the Hanford Redox and Purex plants. It consists of facilities for fuel dissolution, solvent extraction, and processing of waste effluents from the plant. It was designed to handle fuel from the Hanford production reactors. In addition to pilot plant work for the Hanford separations plants, it has also been used for recovery and processing of various fission product materials, especially strontium and cerium. It is presently in an inactive status and could be recommissioned for use in fuel reprocessing operations. However, the facility is designed for contact maintenance, and the cells are not designed for easy and complete decontamination.

EXISTING PROCESSING EQUIPMENT

Dissolver

A dissolver was installed several years ago in C-cell of the 325A facility for use with irradiated neptunium. This dissolver has a solution operating capacity of about 35 liters. It is fabricated from type 825 Incoloy alloy, it is electrically heated by band heaters on the outside, and cooled by a water coil on the inside. It is equipped with a perforated basket for holding the fuel elements. The offgas system includes a condenser, a demister, a caustic scrubber, and a silver zeolite cartridge. This unit is available for processing fuel at a rate of 40 to perhaps 50 kilograms per day.

Centrifugal Contactor

An 8-stage centrifugal contactor is available for processing fuel at a rate of perhaps 2 kg per day. This contactor was shop made about three years ago, and has been used for programs other than fuel reprocessing until quite recently when it was used for processing about 2.6 kg of power reactor fuel. Other centrifugal contactors have also been used in the hot cell, but at present they are not believed to be salvagable.

Pulse Columns

The only pulse columns which are currently available for processing hot material are in the Hot Semiworks. These columns range in size from about 4 to about 12 inch diameter. Since this facility has not been operated for several years, these columns would require extensive checkout and maintenance before they would be operational. Modification might be required to assure criticality safety for power reactor fuels. Installation of new pulse column capability will not be difficult either at Hot Semiworks or elsewhere because of the extensive experience at PNL in designing and operating pulse columns.

SHIPPING AND RECEIVING

Fuel elements can be received in 324 from truck casks and loaded into a water basin. From this location, they could be transferred into the airlock and a shear could be installed in the hot cells to convert the fuel elements to chopped pieces suitable for dissolution. The solid pieces of fuel element could be loaded into a second cask and transported to 325 for dissolution or they could be transported to a dissolver in the 324 hot cells if one were established for this purpose.

Process solutions can be transported between facilities in three types of casks which are currently available. Casks are available with about 8 inches of lead shielding and volumes of either 200 or 300 gallons. A 500 gallon cask has about 4 1/2 inches of lead shielding. The 200 and 500 gallon casks can be handled in any of the facilities while the 300 gallon cask can be handled in the 324 facility or the Hot Semiworks but not in the 325A facility. Because of increasing restrictions on shipments of high level process solutions, it is desirable to restrict the shipments to within the 300 Area (between 324 and 325A) if this is possible. Therefore, liquid shipments to or from Hot

Semiworks would be undesirable. Completion of the hot-solution transfer line between 325A and 324 could reduce cask transfers between these two buildings.

Waste solutions are transported from PNL facilities to ARHCO for ultimate disposal. Low level liquid wastes are transported through a collection facility, then by rail tank car to ARHCO. Higher level solutions are transported in the same casks used for process solution transport. Wastes from Hot Semiworks are sent directly to ARHCO storage tanks.

ALTERNATIVE PROCESSING SCHEMES

Kilogram Per Day Level

Existing facilities in 325A are capable of processing up to about 2 kg of irradiated fuel per day. Operations to be performed would be receipt of chopped fuel, dissolution, feed makeup, feed clarification, solvent extraction, uranium-plutonium disposal, high-level waste shipping to 324, and chemical analysis. This capability has recently been demonstrated on approximately 2.6 kg of power reactor fuel. The major problem encountered in the recent run was fouling of the centrifugal contactors by solids in the liquid streams. This problem was overcome by installing high efficiency filters on all inlet lines to the centrifugal contactor. Processing rate for the existing system is approximately 2 kg per day maximum based on the operating flowrate of the centrifugal contactor.

Operating costs for this type of campaign would be approximately $30,000 for a run of up to 10 kilograms in a five day period. This assumes chopped fuel being received in 325A and the high level waste being transported to 324 without concentration in 325A. Although maximum throughput might be as high as 2 kg per day, it is unlikely that actual sustained throughput could be higher than about one-fourth of this.

The principal problem is expected to be maintenance of the centrifugal contactor and uncertainty in the life expectancy of this contactor. A continuing program with this mode of operation should provide for immediate purchase of an additional contactor and replacement components for the contactor. Cost for this equipment would be approximately $50,000. Some process development on feed clarification would be highly desirable and improved equipment for feed clarification will be required. Fuel processing in existing equipment could be started at any time. In fact, 12 kg of chopped PWR fuel is currently being stored in 324 until funding becomes available for reprocessing.

TONNE PER MONTH LEVEL

Processing at a rate of about one tonne per month has received the greatest attention in this study. Emphasis was placed on performing the solvent extraction operation in 325A so that 324 could be devoted exclusively to waste solidification operations. A nominal processing rate of 50 kg per day was selected. With an operating efficiency of 75 percent, this will correspond to a production rate of slightly over one tonne per month.

Three types of contactors were considered for solvent extraction: pulse columns, centrifugal contactors, and mixer settlers. Centrifugal contactors were ruled out because of the uncertainty of operability with feed which may contain solids. The mixer settlers were ruled out because of excessive contact time between the organic phase and the high level aqueous phase. Pulse columns can be operated in 325 even though the headroom in the cells is lower than would be desired for this scale of operation. Adequate space exists to install a set of three columns for the HA-HC operation. The HA and HS columns would be two inches in diameter, and the HC column would be three inches in diameter. The solvent extraction operations to be performed are extraction of the uranium and plutonium from the aqueous phase into the organic phase (HA column), scrubbing fission products from the organic phase (HS column), and then stripping the uranium and plutonium from the organic phase into an aqueous phase (HC column).

Additional columns and other processing equipment could also be installed in the large cell of the 325A facility so that all of the required processing could occur on a continuous basis at a rate of about 50 kilograms per day. The uranium and plutonium from the HC column would be concentrated and acidifi to about 7 molar nitric acid, and the plutonium would be recovered by processing through an anion exchange column. The plutonium could then be shipped to ARHCO in L-10 containers for final purification and storage. The uranium with small amounts of fission products (about 0.1% of the original) would be treated as a waste. The fission product effluent from the HA column would be sent to 324 for use in the waste program studies.

The existing dissolver is possibly large enough to handle the entire throughput of 50 kg per day. Since the dissolver is the capacity limiting equipment, a second dissolver should be fabricated to provide a backup in the event of future failure of the dissolver. Various techniques for feed clarification will be considered, including both centrifugation and filtration. A method of feed clarification will be selected and suitable equipment will be installed. Existing tanks in the facility appear to be satisfactory for feed makeup and waste collection, provided satisfactory scheduling can be obtained for shipping product materials to 324 and waste or byproduct materials to ARHCO.

Some of the principal concerns of this method of operation relate to safe handling of effluent materials, particularly the offgas stream. Some offgas treatment may be required to assure adequate management of tritium and iodine-129. Additional studies will be required to determine the need for such confinement, and the possible locations of equipment to handle offgas treatment. However, no technical or space limitations appear to exist to preclude operating at the nominal level of 50 kg per day.

Preparation for this scale of operation will require extensive modification of the hot cell facility to permit improved criticality control, and it will require fabrication, testing, and installation of the solvent extraction equipment. Operations are expected to require three technicians per shift, seven days per week. This will require training of about six additional people. Training of new personnel will have to begin as soon as funding is authorized so that they will be available as qualified operators when full hot work begins. Major cost elements in preparation for this scale of operation are design and fabrication of equipment, preparation of the hot cell, installation of equipment in the hot cell, and training of personnel. Total setup costs for renovating the facility and testing the equipment with both cold and low activity level solutions are estimated to be $2,000,000. After setup, annual operating costs will be $1,500,000.

Operations at a throughput of 50 kg per day or higher will cause many of the same problems faced by industry in operating commercial plants. Frequent contact with prospective commercial reprocessors will help to identify potential problem areas, and to identify possible solutions to these problems. In particular, we expect to address problems of characterizing solids in dissolver solution or elsewhere in the process, feed clarification, solids dissolution, effluent characterization and control, solvent washing, process control instrumentation, safeguards, accountability, criticality control, spent solvent disposal, and disposal of solvent wash solutions.

A side benefit of the program will be the supply of cladding hulls which will be generated. These hulls can be used to study methods for decontamination or disposal in an existing NFCP program at PNL.

Our work will be performed in hot cells with master-slave manipulators for highly flexible operations. This will permit remote replacement of much of the equipment, changing the routings of process streams, and free choice of the number, type, and order of process steps. Spare columns will be installed for use in case of malfunction of the primary columns, and these additional columns can be used for an additional cycle of solvent extraction to characterize wastes from later cycles which ultimately go to the HLW. The flexible operating arrangement will permit generating HLW under a wide variety of plant operating conditions.

The principal ERDA programs which would be affected by installing a large-scale fuel processing capability in the 325A hot cells are the BER program on promethium production, and the PR program on americium production. Both of these programs use the large ion exchange columns currently installed in this facility. While it might be possible to retain the ion exchange columns, it would not be possible to operate the solvent extraction and the ion exchange facilities simultaneously. Substantial equipment cleanout would be required between solvent extraction runs and an ion exchange run. We assume that the NFCP effort is a higher priority and will have as much of the facility as is required. Actual priorities will have to be arranged by ERDA-HQ.

For planning purposes, we assumed that ARHCO could accept the plutonium recovered in our processing, and would accept liquid waste on the same basis as in the past. Because of change in waste handling policies over the past few years, we would need to review this with ARHCO early in the program. However, we are waiting for an expression of NFCP interest before beginning the review with ARHCO.

Tonne Per Week Level

An increase to a production rate of one tonne per week would require installation of larger solvent extraction equipment. Because of the limited height in the 325A facility, it would be impractical to install columns much larger than the proposed 2 inch main extraction columns. Larger columns, perhaps up to four inch diameter, could be installed in 324 where substantially more headroom is available. Although space does exist in 324 for such columns to be installed, the operation of these columns would not be compatible with the proposed waste solidification program for which the waste is needed. Operations with flammable solvents are not easily integrated with the high-temperature equipment used for waste solidification. Fuel dissolution, which is more compatible with the waste solidification equipment, could also be performed in 324. In addition to the safety considerations with flammable solvents, the addition of dissolution and solvent extraction capabilities to 324 would occupy space which could otherwise be used for waste solidification operations.

An alternate method of approaching one tonne per week would be to dissolve fuel in 324 and do solvent extraction in 325A. In the 50 kg per day case, the dissolver capacity in 325A is limiting. The solvent extraction capacity might approach 100 kg/day, and an increase in column size to 2 1/2 inches might provide a 150 kg/day capacity. This appears to be the maximum solvent extraction capacity in 325A for a single set of columns. Higher throughput could be achieved by operating two sets of columns in parallel. Rates greater than 50 kg per day would appreciably compound problems of criticality control, waste disposal, plutonium recovery, solution transfer, and solvent washing. However, higher rates are not impossible.

Tonne Per Day Level

Operations at a tonne per day level would be roughly equivalent to the Nuclear Fuel Services separation plant in West Valley, New York. This level could conceivably be achieved at Hanford using the Hot Semiworks plant. This facility is currently assigned to ARHCO but is in standby condition and could be reactivated by ARHCO or it could be transferred to PNL and reactivated.

Most of the modification of Hot Semiworks would be in the head end operations. New equipment, and probably a new building, would be required for fuel receipt, chopping, dissolution, and offgas treatment. Column modifications may also be required because of the higher fissile content of power reactor fuels. The additions to convert this facility for power reactor fuels at the tonne per day level would be very similar to the head end of the NFS plant. The costs for refurbishing this facility have not been estimated with any great accuracy but are believed to be easily in excess of $10,000,000 and perhaps more like $100,000,000.

Operation of this facility would require at least five operating people per shift, together with radiation monitors and maintenance personnel. Because of the contact maintenance nature of the facility, the ability to operate on a sustained basis is rather questionable unless equipment is designed very carefully and tested thoroughly.

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ASSUMPTIONS USED ON STUDY

1. Fuel will be available from North East Utilities Corp. or others at no cost.
2. Shipment of 400 kg fuel per cask will be made at a cost of $7,000 per shipment.
3. Shearing will not cause a significant risk of zirconium fires.
4. Hulls will be transferred to another ERDA program.
5. Recovery of tritium, iodine, and krypton is not required.
6. Dissolver solids can be added to HLW or discarded separately.
7. Satisfactory equipment for feed clarification will not require development effort.
8. No major additions to offgas system will be required outside of cells.
9. Satisfactory accountability procedures can be established.
10. HAW can be transferred to 324 Bldg. by existing 200 gal. cask or new pipeline.
11. Overheads from U-Pu concentrator can go to 300 Area radwaste system.
12. ARHCO will accept partially decontaminated U stream as waste in existing 500 gal. casks.
13. ARHCO will accept Pu as product in L-10 containers.
14. Operations will not require extensive additional safeguards.
15. No new SAR is required for 325 Bldg.
16. EIS is not required.
17. Personnel exposure will not be limiting.
18. NFCP has highest ERDA priority in 325A and 324.

POTENTIAL WASTE SOURCES FOR SOLIDIFICATION PROGRAM

Requirements

1. Low-additive Purex process.
2. Power reactor fuels for correct fission product ratio to inerts.
3. Minimum rate of 10 tonnes per year for demonstration runs.
4. Cooling time of one to three years out of reactor preferred.
5. Liquid waste transportable to versatile solidification equipment.
6. Full program including solidification to be operational within 18 months.

Evaluation of Potential Sources

Nuclear Fuel Services

Accumulated waste fails to meet 1, 4, and 5. Waste is neutralized, so no longer low additive. Long cooled. Liquid waste not transportable.

Future waste fails to meet 1, 5, and 6. Uses iron cans to move chopped fuel to dissolver. Not operational within 18 months. No available solidification equipment.

Allied General Nuclear Services

Fails to meet 1, 5, and 6. Use gadolinium soluble poison. Not operational within 18 months.

No available solidification equipment.

Savannah River Plant

Fails to meet 2, 5, and 6. Not power reactor fuels. Liquid waste not transportable. No available solidification equipment.

Idaho Falls

Fails to meet 1, 2, and 5. Salting used in some processing.

Not typical power reactor fuels.

No versatile solidification equipment.

Hanford - PNL

With proposed program, meets all requirements. In fact, fully operational 12 months after authorization.

POWER REACTOR FUEL PROCESSING POTENTIAL AT PNL

Processing Rate kg per Day	Location, Building	Lead Time	One-Time Cost, $k	Annual Oper. Cost $k	Remarks
1-2	325A	0	50	750	Useful for product evalu-ation and cladding studies
30-50	324 & 325A	9 mos.	2000	2000	Provides full-level waste solidification demonstratio
150-200	324(&325A)	12 mos.	4000	5000	Provides longer full-level waste solidification demons-tions.
750-1000	HSW	3 yr.	>10,000	7000	Higher rate than required?