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**High-Level Waste Program
Progress Report for October 1, 1978,
through March 31, 1979**

R. E. Blanco
A. L. Lotts

MASTER

OAK RIDGE NATIONAL LABORATORY
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NUCLEAR FUEL AND WASTE PROGRAMS

HIGH-LEVEL WASTE PROGRAM PROGRESS REPORT
FOR OCTOBER 1, 1978, THROUGH MARCH 31, 1979

Program Manager: R. E. Blanco

Program Director: A. L. Lotts

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HIGHLIGHTS

1. Waste Management Analysis for Nuclear Fuel Cycles

Development of a low-level waste management analysis computer code capable of being used alone or as a component of an overall waste system analysis program has been initiated.

As part of an ongoing update of the ORIGEN code, models of PWRs fueled with denatured ^{233}U and denatured ^{235}U in ThO_2 are being developed.

Information on waste management for 18 alternative fuel cycles has been assembled for use by NASAP.

Documentation of experimental and calculational work done for the assessment of partitioning and transmutation is progressing, while the conceptual design, cost estimation, and risk analysis of partitioning and waste disposal facilities are scheduled for completion in September 1979.

2. Fixation of Waste in Concrete

Assuming diffusion control, leach studies with FUETAP concretes indicate effective diffusivities in the order of $\leq 10^{-17}$, $\leq 10^{-13}$, and $\leq 10^{-10}$ cm^2/sec for plutonium, cesium, and strontium, respectively.

Hydrogen and oxygen combine rapidly at ambient temperature in the presence of FUETAP concrete.

3. Study of Ceramic and Cermet Waste Forms

A spray calciner is currently being fabricated and installed in a recently acquired trailer laboratory. Its successful operation is a key in the transition from batch to continuous cermet processing.

New cermet sintering techniques have been developed which drastically reduce the sintering temperatures and times required to achieve satisfactory densification.

The radioisotope volatility losses observed during the processing of actual wastes to form cermets were very low, as had been expected from previous results of radiotracer studies.

Conversions of actual SRP acid and dried sludge wastes to cermets have been performed. These samples, along with the cermet sample formed from NFS Acid Thorex waste, are currently undergoing characterization.

A series of cermet leach tests conducted by PNL have been completed, and the samples are currently being examined at ORNL. While complications were encountered during these tests, the results indicate that cermets exhibit promising leach resistance and durability.

Program restructuring, at the request of SRL, has accelerated the initiation of economic, engineering, and safety studies while de-emphasizing cermet process development and the characterization of the resulting cermets, particularly regarding actual waste studies.

1. WASTE MANAGEMENT ANALYSIS FOR NUCLEAR FUEL CYCLES
(Activity No. AG 30 20 01 0; ONL-WH01; ON 1.1.3.2 AG)

J. O. Blomeke, C. W. Alexander, A. G. Croff, B. C. Finney,
C. W. Kee, D. W. Tedder, and J. W. Wachter

1.1 Summary

This program is divided into three principal tasks: (1) waste management analysis, which consists of the development of data bases and computer codes suitable for the analysis of waste management systems; (2) special projects, which take the form of responses to generally specific but unscheduled requests from DOE for technical data and analyses pertinent to waste management; and (3) an assessment of actinide partitioning and transmutation, which is currently in a final phase of cost-risk/benefit determination.

During this report period, Teknekron, Inc. prepared a package of data and descriptive material for use by EG&G-Idaho in the preparation of a contingency plan for the receipt of commercial low-level wastes at DOE burial grounds. Teknekron also submitted a program plan for the development of an overall waste management analysis computer code. Work at ORNL consisted of modifications to our existing waste projection codes to permit a more rigorous accountability of spent-fuel inventories, and of updating the ORIGEN code from the standpoint of its nuclear data libraries and its capabilities for handling alternative fuel cycle PWRs. Special projects that were undertaken included the preparation of waste management information for inclusion in the Technical Data Base of the NASAP final report, and of compilations of information on waste characteristics and constituents that are potentially of beneficial use. The partitioning-transmutation effort was devoted to (1) the documentation of previous experimental work; (2) the documentation of actinide transmutation calculations for PWRs; (3) the conceptual design of a shipment cask for spent fuel containing recycled actinides; and (4) the direction and coordination of the conceptual design of fuel cycle partitioning facilities, and risk analyses of those facilities and a geologic waste repository.

1.2 Waste Management Analysis

1.2.1 Low-level waste management analysis (C. W. Alexander)

Low-level waste management analysis has been primarily directed toward the planning, organization, and development of a computerized LLW system analysis capability. The bulk of this effort has been subcontracted to Teknekron, Inc., and to date, Teknekron has prepared a program plan for development of a computerized waste management systems analysis tool and has supported EG&G-Idaho in the development of a low-level waste (LLW) contingency plan.

The program plan was requested in an attempt to lay the foundation for the orderly and logical development of an overall waste management analysis code. This activity was designed to be a multiyear effort requiring site interactions, extensive surveys of operational experience and data, interfaces between agencies, and the use (wherever possible) of existing codes. The first year will be dedicated to LLW management analysis and should result in a code suitable for use in LLW management systems analysis by the end of FY 1979.

ORNL and Teknekron were also asked by DOE to assist EG&G in the development of a contingency plan for disposal of commercial LLW in DOE burial grounds. Teknekron's contributions were comprised of commercial and institutional waste projections, transportation costs, land burial requirements, scenario development, and definition of events that could lead to the implementation of the plan. This effort was terminated on March 1.

1.2.2 Modifications of waste projection codes (C. W. Kee)

Previous waste projection calculations have been made without accounting for varying decay times of fuel which had been stockpiled due to a shortage of reprocessing capacity. Therefore, all fuel reprocessed was assumed to be the same age at the time of reprocessing. Modifications have now been made to KWIKPLAN to allow for variation in the decay (reprocessing) for each batch of fuel in the stockpile. Calculations have been made with this revised version, and the results were checked for internal consistency and for consistency with previous calculations.

More extensive modifications have been made to WASPR to allow it to use the new KWIKPLAN output and to properly decay the fuel as it is reprocessed from the stockpile. To more readily verify the validity of the new coding, the first modifications assume this decay occurs after reprocessing. Later modifications will allow WASPR to assume that reprocessing occurs after a variable decay time.

1.2.3 ORIGEN update (A. G. Croff)

The ORIGEN^{1.1} computer code is a versatile tool for calculating the buildup and depletion of isotopes in nuclear materials. This computer code was written in the late 1960s and early 1970s in the ORNL Chemical Technology Division, and at that time the required nuclear data libraries (half-lives, cross sections, fission product yields, etc.) and reactor models (PWR-U, PWR-Pu, LMFBR, HTGR, and MSBR) were developed. The code was principally intended for use in generating spent fuel and waste characteristics (composition, thermal power, etc.) that would form the basis for the study and design of fuel reprocessing plants, spent fuel shipping casks, waste treatment and disposal facilities, and waste shipping casks. Since fuel cycle operations were being examined generically, and thus were expected to accommodate a wide range of fuel characteristics, it was only necessary that the ORIGEN results be somewhat representative of this range. A satisfactory result was obtained by simply adjusting the resonance integrals of the major fissile and fertile species to obtain agreement with a spent fuel composition from an exogenous source.

Soon after the ORIGEN computer code was documented, it was made available to users outside ORNL through the Radiation Shielding Information Center (at ORNL). The relative simplicity of ORIGEN, coupled with its convenient and detailed input and output, resulted in its being acquired by many organizations. Many of these organizations began using ORIGEN for applications that required greater precision in the calculated results than those for which it had originally been intended. These applications were generally much more specific than the early ORNL generic fuel cycle studies. Many of them were environmental impact studies that

required relatively precise calculations of minor isotopes such as ^3H , ^{14}C , ^{232}U , and $^{242,244}\text{Cm}$. The initial responses to these requirements were attempts to update specific aspects of ORIGEN and its data bases;^{1,2,1.3} however, inconsistencies and a large number of different data bases soon resulted from these efforts.

In an effort to remedy these problems, a concerted program was initiated in 1975 to update ORIGEN and its associated data bases and reactor models. The three principal aspects of the code that have been or are being updated are: the ORIGEN computer code itself, the reactor models, and the decay-related data. The update of the ORIGEN code was substantially completed during FY 1978, and the revised version is called ORIGEN2. The features of ORIGEN2 that are substantially different from those of ORIGEN are as follows:

1. ORIGEN2 is variably dimensioned to minimize the amount of computer space required.
2. All input to ORIGEN2 is "free-format" (i.e., parameters can appear anywhere on a card as long as they are in the proper order and of the proper type).
3. The user can now specify more general and detailed fuel cycle flowsheets, including recycle calculations.
4. The number of related or unrelated calculations that can be performed in a single job is only limited by the available computer time.

ORIGEN2 is currently in routine use within ORNL, and a user's manual is currently in preparation. A summary report on ORIGEN2 and its data bases will be initiated during this fiscal year.

The largest update activity concerns the "reactor models" used in ORIGEN2. A single reactor model update (e.g., an enriched-uranium-fueled PWR) includes the following data:

1. new cross sections (about 230) for actinide, fission product, and activation product nuclides;
2. new neutron-spectrum-averaged fission product yields for about 850 fission products from six fissile actinide isotopes;

3. burnup-dependent cross sections for the major actinide nuclides (e.g., ^{235}U , ^{239}Pu);
4. new values for the ORIGEN flux parameters THERM, RES, and FAST;
5. parameters related to the activation of fuel assembly structural materials outside the active fuel region;
6. recommended initial compositions and amounts of heavy metal in fresh fuel;
7. recommended initial metal compositions and amounts of fuel assembly structural materials; and
8. recommended minor constituent concentrations for both the fuel and structural materials.

The first five of the above items result from multigroup neutron spectrum and depletion calculations for each fuel type in each reactor.^{1.4} The details of the calculations depend on the reactor being considered. The principal sources of cross-section data have been the ENDF/B compilations.^{1.5,1.6}

During this report period the reactor models for the U-Pu cycle PWRs and BWRs were completed and documented.^{1.7} Work is currently in progress on alternative fuel cycle PWRs. Reactor physics calculations have been completed and ORIGEN2 calculations are under way for the following alternative fuel cycle reactor models:

1. denatured ^{233}U -enriched thorium-fueled PWR;
2. plutonium-enriched thorium-fueled PWR; and
3. an extended-burnup (50,000-MWd/MTHM), ^{235}U -enriched uranium-fueled PWR.

Reactor physics calculations are well under way for a PWR enriched with self-generated, denatured ^{233}U and denatured ^{235}U in ThO_2 . Reactor physics calculations have just been initiated for a variety of standard and alternative fuel cycle LMFBRs.

The final aspect of the update effort concerns decay-related data. Decay-related data include data concerning (1) the half-life, decay branching ratios, and decay heat; and (2) the photons resulting from

radioactive decay events. The first type of decay-related data is contained in the ORIGEN2 decay library, which includes:

1. half-life;
2. the probability of beta decay to an excited state;
3. the probability of (positron + electron capture) decay to an excited state;
4. the probability of (positron + electron capture) decay to a ground state;
5. the probability of alpha decay;
6. the probability of the internal transition decay of an excited state;
7. the average, recoverable energy per decay event;
8. the probability of spontaneous fission decay;
9. the probability of decay via emission of a beta particle and a neutron (i.e., (β ,n) decay);
10. the natural abundance; and
11. the RCG values for both air and water.

The second type of decay-related data is contained in the ORIGEN2 photon library and includes:

1. gamma rays and x-rays resulting directly from radioactive decay,
2. bremsstrahlung (photons from the slowing down of beta particles),
3. gamma rays from spontaneous fission events and from the spontaneous fission products, and
4. gamma rays from (α ,n) reactions.

The decay-related information required for the update came from two principal sources: ENDF/B-IV^{1.5} and ENSDF.^{1.8,1.9} In the current update, the decay-related information for 427 of the most important nuclides in ORIGEN2 was updated using information from ENSDF. The decay-related data for the remaining radioactive isotopes (ca. 600), which are mostly short-lived fission products, were updated using information from ENDF/B-IV. The results of this update are complete and have been documented.^{1.10}

1.3 Special Projects

1.3.1 NASAP waste management data base (B. C. Finney and D. W. Tedder)

An AFCEP (Alternative Fuel Cycle Evaluation Program) Characterization Working Group was organized with representatives from various government sites and industry to generate a technical information data base that can be used to evaluate and compare 18 proliferation-resistant fuel cycles. The AFCEP data base will be incorporated into the NASAP (Nonproliferation Alternative System Assessment Program) Technical Information Data Base and will serve as the source material for writing a chapter on fuel cycle operations in the NASAP final report. Eighteen fuel cycles are being evaluated (Table 1.1). An example of the basic outline for each fuel cycle for the AFCEP Technical Information Data Base is as follows:

- 1.1 Fuel Cycle Description and Flowsheet (including fissile mass flows)
 - 1.1.1 Fuel fabrication
 - 1.1.2 Reactor
 - 1.1.3 Fuel reprocessing
 - 1.1.4 Waste conditioning and disposal
 - 1.1.5 Transportation
- 1.2 RD&D and Technology Status and Feasibility Study
 - 1.2.1 Fuel fabrication
 - 1.2.2 Reactor
 - 1.2.3 Fuel reprocessing
 - 1.2.4 Waste conditioning and disposal
 - 1.2.5 Transportation
- 1.3 Safety, Licensing, and Environmental Data
 - 1.3.1 Fuel fabrication
 - 1.3.2 Reactor
 - 1.3.3 Fuel reprocessing
 - 1.3.4 Waste conditioning and disposal
 - 1.3.5 Transportation
- 1.4 Cost Data
 - 1.4.1 Fuel fabrication
 - 1.4.2 Reactor
 - 1.4.3 Fuel reprocessing
 - 1.4.4 Waste conditioning and disposal
 - 1.4.5 Transportation

Table 1.1. NASAP fuel cycles that are being evaluated

Case number	Fuel cycle
1	LWR — once-through
2	LWR — modified once-through
3	LWR — U/Pu recycle (reference case)
4	LWR — U/Pu spiked recycle
5	LWR — DU(3)/Th recycle ^a
6	Backfit prebreeder
7	Advanced breeder
8	Prebreeder, Shippingport type 1
9	Breeder, Shippingport type 1
10	HWR DU(5) — once-through ^b
11	LMFBR U-Pu coprocessed recycle
12	LMFBR U-Pu spiked recycle
13	LMFBR U-Pu/Th spiked recycle
14	LMFBR Th-Pu/Th spiked recycle
15	LMFBR DU(3) Th/Th recycle
16	HTGR DU(5) — once-through
17	HTGR DU(5)/DU(3) — Th
18	GCFR U-Pu/Th spiked recycle

^aDU(3) denotes denatured ^{233}U .

^bDU(5) denotes denatured ^{235}U .

We were asked to provide the technical information on waste characterization and conditioning, environmental and licensing, and costs for the reactors and the integrated (fabrication and reprocessing) facilities; LASL is responsible for shallow-land burial; Sandia for waste transportation; and SAI/ONWI for the repository. We were also given the responsibility of compiling all the above information into a single document suitable for use by NASAP.

Work on the task was begun in early January, and a draft document of the ORNL information was sent to DOE and members of the AFCEP Characterization Working Group the end of January. A revised document of the ORNL information was completed at the end of February.

SAI/OR has been contracted to compile the AFCEP Technical Information Data Base which is scheduled to be sent out for industrial review on April 2, 1979. The NASAP final report also to be prepared by SAI/OR is scheduled to be completed on May 1, 1979, and the report is to be submitted to the President and Congress by December 31, 1979.

1.3.2 Potentially beneficial waste constituents (J. O. Blomeke and C. W. Alexander)

During this report period two requests were received from DOE for information on the characteristics of high-level waste and spent fuel, and on the potential uses of various waste constituents. This information, largely based on ORIGEN calculations, was supplied in the form of figures and rather extensive tables depicting the heat-generation rates, radioactivity levels, type and energy of decay, potential uses of specific fission products and actinides, and the status of technology for beneficial purposes.

1.4 Partitioning and Transmutation

1.4.1 Documentation of experimental programs (D. W. Tedder)

All experimental work relating to actinide partitioning was terminated at the end of FY 1978. Since that time, documentation efforts have continued both at ORNL and off-site (i.e., ANL, RFP, and ICPP). The following reports are in progress and due to be completed in the near future:

ORNL

- D. W. Tedder, B. C. Finney, and J. O. Blomeke, Waste Treatment Facilities for the Partitioning of Actinides from LWR Reprocessing and Refabrication Plant Wastes, ORNL/TM- (in progress).
- C. W. Forsberg, Computer Simulation of Displacement Cation Exchange Chromatography Separation of Trivalent Actinides and Lanthanides with Initial Benchmark Experiment, ORNL/TM-6738 (in progress).
- F. M. Scheitlin and W. D. Bond, Recovery of Pu from HEPA Filter Media by Continuous Leaching in a Packed Column with Electrolytically Produced Ce(IV) Nitrate and Nitric Acid, ORNL/TM-6802 (in progress).
- C. W. Forsberg, The Elution Sequence of Neptunium and Plutonium from a Cation Exchange Chromatography Column Eluted with Diethylenetriaminepentaacetic Acid (DTPA), ORNL/TM-6737 (in progress).
- C. W. Forsberg, Effects of Bidentate Solvent Extraction Chemicals on the Distribution Coefficients of Rare Earths on Cation Exchange Resins, ORNL/TM- (in progress).

ANL

- E. P. Horwitz et al., The Recovery of Actinides from TBP-Na₂CO₃ Scrub Waste Solutions: The ARALEX Process, ANL- (in progress).

RFP

- G. H. Thompson et al., Actinide Recovery from Combustible Waste: The Ce(IV)-HNO₃ System, RFP- (in progress).

ICPP

- L. D. McIsaac et al., Study of Bidentate Compounds for Separation of Actinides from Commercial LWR Reprocessing Wastes, ICP-1180 (December 1978).

In addition, a number of papers will be presented at the Actinide Separation Symposium, ACS Pacific Chemical Conference, Honolulu, Hawaii, April 3-5, 1979. The papers resulting from experimental work that was funded by the actinide partitioning program and to be published by the ACS are as follows:

- D. W. Tedder, B. C. Finney, and J. O. Blomeke, "Actinide Partitioning Flowsheets," ORNL.
- E. P. Horwitz et al., "The Extraction of DBP and MBP from Actinides: Application to the Recovery of Actinides from TBP- Na_2CO_3 Scrub Solutions," ANL.
- L. D. McIsaac et al., "Flowsheet Development Work at the Idaho Chemical Processing Plant for the Partitioning of Actinides from Acidic Nuclear Waste," ICPP.
- W. D. Bond and R. E. Leuze, "Removal of Americium and Curium from High-Level Wastes," ORNL.

1.4.2 Transmutation calculations (J. W. Wachter)

The program to evaluate the feasibility and incentives for partitioning-transmutation (P-T) as a waste management concept requires knowledge of the radionuclide characteristics of the P-T fuel cycle. Calculations have been carried out at ORNL and at SRL to examine the nuclear characteristics of a reference LWR fuel cycle which does not recycle transplutonium actinides and a fuel cycle which uses P-T. The radionuclide mass balances calculated from these model fuel cycles have been used to explore the technical and economic implications of a P-T fuel cycle with respect to the reprocessing, transportation, and fabrication components of the fuel cycle.

In conventional plutonium utilization schemes, the plutonium is recovered from the spent fuel, converted to the oxide, and mixed with either natural or depleted uranium dioxide as a diluent. The proportion of each oxide in the mixture is adjusted so that the resulting mixed oxide (MOX) fuel has the same nuclear reactivity as the enriched uranium usually burned in the reactor. Consequently, the amount and nuclear reactivity of the plutonium recovered from the spent fuel determine the number of fuel pins which can be fabricated from the recycle MOX and, hence, the fraction of the next reload which is occupied by the recycle material. When coprocessing techniques are used during spent fuel reprocessing to blend the uranium and plutonium in the correct proportions for the next recycle MOX fuel, a significant amount of neptunium

is carried through as well, but calculations indicate that the nuclear poisoning by the neptunium can be compensated for by increasing the plutonium content and thereby decreasing the number of recycle fuel rods. If both coprocessing and actinide partitioning are employed, so that all the actinides are carried through to the MOX fuel, the required fissile plutonium content increases greatly. Calculations have been made by both the ORIGEN code at ORNL and the GLASS code at SRL which indicate that only a few recycles of the actinides are possible before the plutonium content of the rods becomes so great that the power peaking in the pins exceeds the tolerance level for LWRs.

The power peaking problems in the MOX pins can be completely eliminated if the recycled actinides are distributed uniformly throughout the entire reactor reload since then the transuranium (TRU) actinides are diluted by their homogeneous dispersal in the enriched uranium dioxide fuel. However, this method is not desirable because of the excessive cost of fabricating the entire reactor core under the radioactivity standards required of plutonium fuel fabrication. Survey calculations using the ORIGEN code were performed to determine the smallest fraction of the reload fuel in which the TRU actinides could be dispersed while still permitting MOX assemblies to replace enriched uranium assemblies in an LWR. This fraction was found to be approximately 33%.

The neutronic characteristics of the recycle fuel were estimated using GLASS calculations. A calculational model comprising a MOX assembly placed between two enriched uranium assemblies was used. The uranium enrichment of the MOX assemblies was determined by specifying that the ending reactivity of the MOX rods be constant from recycle to recycle. This was necessary in order to take account of the changing reactivity drop-off of the MOX rods over the several recycles. This change in reactivity of the rods during irradiation in the reactor decreases as the waste actinide content of the rods builds up in successive recycles.

Based on these calculations, a model fuel cycle was devised in which the elements were distributed throughout one-third of the reactor reload. The reference nuclear fuel cycle (no P-T) is one employing uranium/plutonium coprocessing followed by fuel fabrication and insertion of the

recycled fissile material into the reactor at the end of two years. The reactor is assumed to be a PWR producing 1250 MW(e) at an 80% capacity factor, so that the fuel entering the processing facility has produced 1.0 GW-yr of electricity. Commercial enriched-uranium reactors are assumed to use recycled uranium, so that the effects of the concentration of the minor isotopes of uranium by the enrichment facilities are taken into account in the enriched-uranium fuel.

Material balances were calculated for each of the actinide elements in the nuclear fuel cycle, account being taken of the necessary cooling periods and delays between processing steps. The reprocessing parameters and the losses in fuel fabrication were chosen to be representative of attainable practice. Changes in the radionuclide inventory and in the nuclear reactivity of the fuel were taken into account in determining the reactor charge at the start of each cycle. This information was made available to other facilities as a basis for assessing the feasibility of the partitioning-transmutation concept.

A technical report detailing the GLASS code calculations performed at SRL is in the final stages of publication at SRL. A draft version of a summary report describing all transmutation calculations has been written and distributed for internal comment at ORNL.

1.4.3 P-T cask design (C. W. Alexander)

A major fuel cycle impact resulting from the introduction of P-T is the change in transportation requirements. The enhanced recovery and recycle of the waste actinides require, after a few irradiations, shipping casks that have substantial neutron shielding in addition to extensive gamma shielding. The neutron shielding problem exists for both fresh and spent fuel. The heat load of a P-T assembly is also very large due to the presence of curium isotopes. Few shipping cask designs exist that even approach the requirements for shielding P-T fuel, and these designs are primarily for breeder fuel, which is both shorter in length than the PWR assemblies considered in this study and less neutron-active.^{1.11,1.12,1.13} However, these designs for FBR fuels provide an excellent basis for the conceptual design of a P-T shipping cask.

Spent P-T fuel, 1.5 years after reactor discharge, was assumed to have a neutron emission rate of 5×10^{11} neutrons/sec-assembly and a photon emission rate of $\sim 2 \times 10^{16}$ photons/sec-assembly. Utilizing the breeder cask designs, the spent P-T fuel radiation characteristics, and a somewhat non-optimized selection process, the search for adequate shielding materials was initiated using the one-dimensional shielding code, ANISN.^{1.14} Properties other than the individual shielding characteristics were considered in the selection process. Some of these properties were weight, thermal conductivity, thermal and chemical stability, abundance, cost, and ease of fabrication. Several ground rules for acceptance were also established. These included the requirement that the dose six feet from the cask surface be no greater than 10 mrem/hr under operating conditions and that the dose three feet from the cask surface be no greater than 1000 mrem/hr under accident conditions. The cask had a 100-metric-ton limit, fully loaded, and could be no greater than 11 ft in diameter. The cask must carry six assemblies, be passively cooled, and contain no water.

The final materials selected were aluminum, SS-304, boron carbide, lithium hydride, uranium, and copper. The aluminum was selected for the fuel basket material and the external heat rejection fins. SS-304 was selected primarily for its structural strength and integrity. It also has some value in gamma shielding. Boron carbide was selected principally for its neutron absorption capability and almost total lack of secondary gamma radiation. Lithium hydride was selected for its high hydrogen density (moderating capability) and its overall density. Uranium was selected for its gamma shielding qualities. Finally, copper was selected as a matrix material for one boron carbide layer and for internal heat transfer fins. All the above materials have a reasonable natural abundance; however, there are some potential problems. Four potential problem areas are: (1) the extreme reactivity of lithium hydride, (2) hydrogen embrittlement of uranium, (3) lack of large-scale production facilities for some of the materials, and (4) actual fabrication of the cask.

The shield design, Fig. 1.1, is a multi-layer configuration which, generally, helps to reduce the weight and alleviates spectral hardening. The shield design is capable of holding six spent P-T fuel assemblies with an operating condition dose of ~ 1.4 mrem/hr. ANISN runs were made with the lithium hydride zone absent, which was assumed to be a possible worst accident. Accident condition dose rates were on the order of ~ 70 mrem/hr. The weight of the shield over the actual assembly length (405.9 cm) is ~ 84.0 metric tons, fully loaded, but this does not take into account the weight of the end pieces.

Recent calculations indicate that fifth-recycle spent P-T fuel (twenty-five years into a P-T nuclear strategy) has a neutron emission rate of $\sim 5 \times 10^{10}$ neutrons/sec-assembly. Calculations beyond the fifth recycle have not been made, but the shield is sufficiently over-designed to compensate for the anticipated growth in the neutron emission rate in recycles beyond the fifth. Two-dimensional DOT^{1.15} runs will be necessary to design the end pieces and more accurately optimize the circumferential shield. Coupled with the overall conservatism in the dose rates, the shield design should be able to meet the ground rules regardless of any perceived inaccuracies in the ANISN cross-section data.

1.4.4 Economic analysis (D. W. Tedder, B. C. Finney, and Ralph M. Parsons Co.)

An economic analysis is being carried out by the Ralph M. Parsons Company. This study includes an analysis of both the fuel reprocessing plant (FRP) and the fuel fabrication plant (FFP) waste treatment facilities (WTFs). The WTFs treat all actinide wastes that are produced during fuel recycle, recover the actinides, and recycle them back to the main reprocessing and fabrication facilities. In addition, Ralph M. Parsons Co. will prepare a cost estimate for a shipping cask that is specially designed to transport the transmutation fuel (see Sect. 1.4.3).

A conceptual study will be completed on both the WTFs and the shipping cask. The WTF study includes: completion of project and instrumentation drawings (P&IDs), equipment sizing and inventory calculations, shielding requirement calculations, building and process layouts, HVAC analysis, site services and location, construction material estimates,

Fig. 1.1. Conceptual shielding design for spent P-T fuel.

and cost estimates. This information will be presented as a final report at the end of FY 1979.

At this time, the P&IDs are nearing completion, and Ralph M. Parsons Co. has begun inventory calculations for equipment items listed on the P&IDs. The following P&IDs are either completed or are nearing completion after revision:

SK-MP-1	Conceptual P&ID, Rev. 2, FFP-WTF Cation Exchange Chromatography
SK-MP-2	Conceptual P&ID, Rev. 1, FFP-WTF CMP Extraction, Part 1
SK-MP-3	Conceptual P&ID, Rev. 1, FFP-WTF CMP Extraction, Part 2
SK-MP-4	Conceptual P&ID, Rev. 1, FFP-WTF Nitric Acid Recovery
SK-MP-5	Conceptual P&ID, FFP-WTF Salt Waste Treatment, Part 1
SK-MP-6	Conceptual P&ID, FFP-WTF Salt Waste Treatment, Part 2
SK-MP-7	Conceptual P&ID, Rev. 1, FFP-WTF Solid Alpha Waste Treatment
SK-MP-8	Conceptual P&ID, FRP-WTF Hulls Leaching and Dissolver Solids Handling
SK-MP-9	Conceptual P&ID, FRP-WTF Cation Exchange Chromatography
SK-MP-10	Conceptual P&ID, FRP-WTF High-Level CMP Extraction
SK-MP-11	Conceptual P&ID, FRP-WTF HLLW CMP Recycle
SK-MP-12	Conceptual P&ID, FRP-WTF Low-Level TBP and CMP Extraction
SK-MP-13	Conceptual P&ID, FRP-WTF Low-Level CMP Recycle
SK-MP-14	Conceptual P&ID, FRP-WTF TBP Recycle

SK-MP-15	Conceptual P&ID, FRP-WTF Nitric Acid Recovery
SK-MP-16	Conceptual P&ID, FRP-WTF Salt Waste Treatment, Part 1
SK-MP-17	Conceptual P&ID, FRP-WTF Salt Waste Treatment, Part 2
SK-MP-18	Conceptual P&ID, FRP-WTF Alpha Waste Treatment
SK-MP-100	Instrumentation Detail

Parsons has transmitted all inventory calculations for the FFP-WTF to both ORNL and SAI, and equipment lists for cost estimation are either completed or nearing completion for P&IDs SK-MP-1 through SK-MP-7. Also, for the FFP-WTF the hot-cell ventilation requirements have been preliminarily outlined and the necessary reference material has been obtained. Detailed equipment information has been procured on solvent extraction, centrifugation, and process solution pumping.

Currently, Ralph M. Parsons Co. expects to complete the line and equipment sizing for both waste treatment facilities by April 6. Although they have fallen slightly behind the original schedule, it still appears probable that all of their work will be completed by the end of the fiscal year.

1.4.5 Fuel cycle facility risk analysis (A. G. Croff and SAI-Palo Alto)

One of the impacts of partitioning-transmutation (P-T) is expected to be an increase in risk resulting from the increased amount of processing of the actinide-bearing wastes and the increased amounts of shorter-lived actinides in the nuclear fuel cycle. ORNL has contracted with Science Applications, Inc. (SAI) to perform a preliminary risk analysis of fuel cycle facilities and operations that would be significantly affected if P-T were implemented. This information will then be fed into the final, overall cost-risk/benefit analysis. This work began during the summer of 1978 and will be completed by October 1979. Because of this schedule, work is presently at an intermediate point and results are not available

at this time. The following paragraphs will briefly outline the approach being taken and progress to date.

Earlier work at ORNL determined that the three fuel cycle operations expected to be significantly affected if P-T were to be implemented are spent fuel reprocessing, mixed-oxide fuel fabrication, and fresh-fuel and spent-fuel transportation. The general approach taken in the fuel cycle risk analysis was to define two plutonium-recycle PWR fuel cycles that are similar in all respects except that one includes provisions for partitioning (P-T cycle) and one does not (reference cycle). Both cycles assume coprocessing flowsheets for the reprocessing plant and colocated reprocessing and MOX fuel fabrication plants. Both the fuel reprocessing plant (FRP) and the fuel fabrication plant (FFP) are designed so that the extra processes required for partitioning are in a physically separate, but nearby, facility. These facilities are called waste treatment facilities (WTFs). This concept results in the operations that are performed in the FRP and FFP being nearly identical; thus the risk analysis for these facilities is similar in both the reference and P-T fuel cycles.

The scope of the risk analysis being done by SAI is as follows:

1. accidental risk from the FRP, FFP, FRP-WTF, and FFP-WTF;
2. dose from routine emissions from these four facilities;
3. an estimate of occupational risk from these facilities; and
4. an estimate of the routine and accidental dose from fresh and spent fuel transportation.

The accidental risk analyses for the four fuel cycle facilities and transportation are based on fault-tree analyses of the systems in question using methods previously developed by SAI.^{1.16,1.17,1.18} The accidental risk analyses for the FRP, FFP, and reference fuel transportation basically involves a straightforward extension of SAI's previous work on reprocessing, fabrication, and transportation risks. The accidental risk analyses for the two WTFs and P-T fuel transportation are somewhat more complex since they must be based on conceptual designs alone and there has been no experience with the facilities and operations involved. The identification of accident sequences and the future probabilities will be largely based on the experience of the analysts and on similarities to existing operations.

Routine doses will be calculated using source terms supplied by ORNL coupled with standard dispersion and dose models. In the case of transportation, the routine dose will be calculated based on the number of shipments, assuming a fixed dose per shipment. Occupational dose will be calculated by assuming a constant, average exposure per employee and basing the facility staffing estimates on the conceptual design study by R. M. Parsons (Sect. 1.4.4).

The net result of this work will be an estimate of the extra short-term risk that would be expected if P-T were to be implemented. These results will be available on both an absolute and a differential basis and will be compared to the costs and benefits of P-T in the final, overall analysis.

At the present time, the status of this work is as follows:

1. FRP: hazards analysis nearly complete, fault trees partially complete from previous work.
2. FFP: hazards analysis begun.
3. FRP-WTF: hazards analysis complete, fault trees under way.
4. FFP-WTF: will be based on a subset of the FRP-WTF analysis.
5. Transportation: risk analysis of spent fuel transportation is complete; transportation of fuel between the FRP and the FFP, between the FFP and the reactor, and of wastes will be based on this analysis.
6. Routine dose: coding modifications to account for more actinides are complete; analysis is under way.
7. Occupational dose: currently investigating the average occupational exposure from operating facilities.

1.4.6 Repository risk analysis (A. G. Croff and Los Alamos Technical Associates)

One of the principal beneficial effects of P-T would be to reduce the long-lived nuclide content of the radioactive wastes isolated in a repository. The purpose of the repository risk analysis is to quantitatively assess the magnitude of this benefit. This work is being performed under subcontract to ORNL by Los Alamos Technical Associates (LATA). The

work being performed is a straightforward application of a radioactive waste repository risk analysis computer code previously developed by Logan et al.^{1.19} This code, called AMRAW, performs a probabilistic risk analysis of a radioactive waste repository using fault-tree analysis.

The general approach taken in the repository risk analysis is to perform two parallel risk analyses, one on a repository containing partitioned (actinide-depleted) wastes and one containing normal wastes. The repository site considered in this analysis is the Los Medanos area in southeastern New Mexico, which is under study as the location of the Waste Isolation Pilot Plant. In this base case, two waste types were defined: Type A and Type B. Type A waste has leach parameters and surface characteristics typical of many immobilized wastes. Type A waste includes high-level, concreted TRU liquid and incinerator ash, iodine, and ^{14}C wastes. Type B waste includes unimmobilized cladding and solid, noncombustible TRU wastes. The Type A wastes are assumed to have a surface-to-volume (S/V) ratio of 0.2 cm^{-1} . For the Type B wastes the S/V ratio is 35 cm^{-1} . This consideration alone will result in the nuclide leach rate from the Type B waste being a factor of $35/0.2 = 175$ greater than that of the Type A waste. The nuclide composition of each of the radioactive wastes was calculated by ORIGEN^{1.20} based on cross-section data supplied by SRL (see Sect. 1.4.2).

The AMRAW computer code will calculate the probabilistic risk from the two waste repositories (with and without P-T) in the base case over a one-million-year time horizon. The AMRAW computer code includes both "slow" release events, such as the intrusion and leaching of the waste, as well as rapid events such as earthquakes and waste exhumation by meteorites and volcanoes. The data used in Logan's original repository risk analysis have been updated to reflect the latest experimental results, particularly with respect to K_d values and the assumed water velocity used during the slow-leach incident.

The results of the repository risk analysis will be a calculation of the expected long-term dose from the repository. The results of this calculation can be expressed in the following terms:

1. radioactivity release (curies),
2. dose rate and dose (rem/yr and man-rem),
3. health effects, and
4. monetary units.

In general, the code produces all of these results during a single AMRAW calculation. These results will be used to compare the performance of the two repositories with each other, with natural background radiation, and with the short-term risks and costs of P-T (see Sects. 1.4.4 and 1.4.5). The risk analysis will indicate which nuclides in the waste control the repository risk in both cases. AMRAW will also be used in its "consequence analysis" mode to calculate the effects of a specific type of repository accident assumed to occur at a specific time. This type of analysis is required to determine the maximum dose rates resulting from a particular accident.

A second major aspect of the repository risk analysis is the sensitivity analyses that will follow the base-case analysis described above. The sensitivity analyses will vary the principal input parameters within wide bounds to determine if data uncertainties are likely to impact the final, overall analysis of the incentives for P-T. Parameters that are likely to be varied in these studies include the leach water velocity, important K_d values, and the waste leach rate/surface area characteristics.

This activity began during the summer of 1978 and is expected to be completed by October 1979. At the present time, changes in AMRAW to accommodate the heavier actinides and the new output tables have been completed and verified. The risk analysis of the base case is currently under way.

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2. FIXATION OF WASTE IN CONCRETE (Activity No. AG 30 20 01 0; ONL-WHO2; ON 1.3.1.1 AG)

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J. H. Paehler, and G. C. Rogers

2.1 Summary

Ceramic solids, which demonstrate much promise as radioactive waste hosts, were prepared from cementitious mixtures containing 10 to 15 wt % simulated radioactive waste. The mixtures were cured for 24 hr at 100 to 250°C and 600 psi. The resulting FUE_{TAP} (Formed Under Elevated Temperatures And Pressures) concretes had a compressive strength of about 30 MPa, a thermal conductivity of 0.5 W/mK, and a permeability in the order of 10^{-5} darcy. The initial leach studies with such solids indicated effective diffusivities in the order of $\leq 10^{-17}$, $\leq 10^{-13}$, and $\leq 10^{-10}$ cm²/sec for plutonium, cesium, and strontium, respectively. Hydrogen and oxygen combine rapidly in the presence of these solids. Thus, canister pressurization due to the radiolysis of the water in solids containing actual waste should be eliminated or at least reduced significantly below that predicted from previous studies.

2.2 Program Objectives

Work is in progress to assess the problems associated with the incorporation of high-specific-activity waste in concrete. More specifically, the objective of this work is to determine the limits of the role that ceramic types of concrete formed under elevated temperatures and pressures (FUE_{TAP} concretes) can play in waste fixation.^{2.1} Other workers^{2.2} demonstrated that concretes formed under conditions of high temperatures (150 to 400°C) and high pressures (25,000 to 100,000 psi) produced solids that showed promise for the immobilization and isolation of radioactive waste. The current ORNL investigation examines the potentialities of FUE_{TAP} concretes (i.e., low-temperature ceramics) formed under less stringent conditions. Initial results showed that hard, durable solids containing up to 15 wt % simulated radioactive sludge

could be made by curing cementitious mixtures for 24 hr at 250°C and 600 psi. Current studies examine:

1. the effect of mix composition and curing conditions on the physical properties of FUETAP concretes;
2. the leachability of strontium, cesium, and plutonium from FUETAP concretes;
3. the use of additives to decrease the leachability of radioactive materials; and
4. the combination of hydrogen and oxygen in the presence of FUETAP concretes.

2.3 Preparation and Testing of FUETAP Concretes

Ceramic solids were prepared by curing various cementitious mixtures (Table 2.1) for 24 to 72 hr at 100 to 250°C and 600 to 1000 psi. In these mixtures, either Type I, III, or high-alumina cement was used. Fly ash, a pozzolanic material, was added to reduce the strontium leachability and react with lime released during the hydration of the cement. An illitic clay was present to fix any cesium present in the radioactive waste. A mixture of hydrated oxides of primarily iron, aluminum, and lead was used to simulate the radioactive waste.

Table 2.1. Example of FUETAP concrete mix

Component	wt %
Cement	27.55
Fly ash	22.50
Illitic clay	8.95
Simulated waste ^a	10.00
Suspending agent	0.40
NaNO ₃	2.60
Water	28.00

^aHydrated oxides of primarily iron, aluminum, and lead.

Type I and Type III cement produced quite similar solids thus Type I Portland cement was used in the curing studies. cement was unsuitable for preparing FUETAP concretes. The density, compressive strength, thermal conductivity, and permeability values were inferior to concretes prepared with Type I cement.

Except for an increase in porosity with temperature, there were no marked correlations of temperature with the physical properties of specimens cured at 100 to 250°C and 600 psi (Table 2.2). The resulting concretes had an average compressive strength of about 30 MPa, a thermal conductivity of 0.50 W/mK, and a permeability in the order of 10^{-5} darcy. Curing at a higher pressure, 1000 psi, caused no significant change in the physical properties. Longer curing periods, 48 or 72 hr, produced solids with 20 to 30% less compressive strength.

Table 2.2. Effect of curing temperature on the physical properties of FUETAP concrete^a

Curing temperature ^b (°C)	Compressive strength (MPa)	Thermal conductivity (W/mK)	Porosity (%)	Permeability (darcy)
c	19.8	0.59	36	9×10^{-5}
100	29.0	0.43	38	3×10^{-6}
160	30.8	0.55	41	1×10^{-5}
180	28.3	0.50	40	1×10^{-5}
250	27.3	0.51	43	2×10^{-5}

^aPrepared with Type I Portland cement and the mix shown in Table 2.1.

^bExcept for the control, specimens were cured at 600 psi for 22.5 hr after an initial 1.5 hr required to heat the specimen to the desired temperature.

^cControl sample. This specimen was cured at ambient temperature and pressure for 28 days.

Durability tests are in progress in which FUETAP specimens are contacted with 10 volumes of either simulated Sandia Type B brine or groundwater for 96 hr at 250°C and 2500 psi to determine if there are any losses to the aqueous phase. Preliminary tests at 217°C and 300 psi showed that losses to the aqueous phase were greater from specimens containing nitrate. About 2.9% of the calcium, 0.07% of the aluminum, and 0.02% of the iron were lost from specimens containing 2.6 wt % NaNO_3 . The calcium loss was 10% greater, and the aluminum and iron 3.5 times greater than the values obtained with specimens containing no nitrate. The first test at 250°C and 2500 psi with a concrete containing nitrate showed calcium losses of $\leq 2.38\%$ and $\leq 0.58\%$ into simulated brine and groundwater, respectively. Additional studies will be made to determine the significance of this loss and the effects of time, curing procedure, and mix composition on the durability of FUETAP concretes.

2.4 The Leachability of FUETAP Concretes

An automatic leach changer was constructed that will accommodate 32 specimens and 4 different leachants (Fig. 2.1). This device is currently being used to determine the leachability of cesium and strontium from FUETAP concrete with the composition listed in Table 2.1. Assuming the leach mechanism is essentially diffusion-controlled, preliminary results indicate effective diffusivity values for cesium of 6×10^{-15} , 9×10^{-15} , and $7 \times 10^{-13} \text{ cm}^2/\text{sec}$ from distilled water, spring water, and Sandia Type B brine, respectively. Under the same conditions, the values obtained for strontium are 6×10^{-11} , 8×10^{-11} , and $2 \times 10^{-10} \text{ cm}^2/\text{sec}$, respectively. Initial data suggest that the strontium leachability may be decreased by the addition of 1 wt % BaO plus 1 wt % Na_2SiO_3 to the concrete during the mix step. The leachability is further reduced if 1 wt % MnO_2 is also added at the same time. Sufficient data have not been collected to estimate the resulting effective diffusivities in these leach tests.

Studies are also in progress to determine the leachability of plutonium from FUETAP concretes. The leach rates are extremely low.

Fig. 2.1. Automatic leach changer.

Assuming diffusion control, the effective diffusivities from specimens containing 5 $\mu\text{Ci } ^{239}\text{Pu}$ are $\leq 1 \times 10^{-19}$ and $2 \times 10^{-17} \text{ cm}^2/\text{sec}$ into spring water and distilled water, respectively. Initial results with a specimen containing 50 $\mu\text{Ci } ^{239}\text{Pu}$ verify this low leachability into spring water. Some of the more recent data suggest that corrosion may also be involved in the distilled water tests. These studies will continue until sufficient data are collected to establish the leach mechanism. In addition, work is in progress to develop a solvent extraction procedure to use in the analysis for plutonium in brine leachants since the current procedure is not effective with such solutions.

2.5 Studies on the Combination of Hydrogen and Oxygen

Results from laboratory studies show that hydrogen and oxygen combine rapidly at ambient temperature in the presence of FUETAP concrete. These high rates suggest that, if the gases produced by the radiolysis of water in actual radionuclide waste solids remain in contact with the concrete, the possibility of excessive canister pressurization should be eliminated or greatly diminished. In these tests, nitrogen containing 4.1% H_2 , 1.7% O_2 , and $\geq 400 \text{ ppm H}_2\text{O}$ was passed through particles of concrete at various temperatures. No measurable water was detected on passing the gas mixture at ambient temperatures over a neat cement paste made with Type I Portland cement. Only 0.1 vol % H_2O was produced when the temperature was elevated to 130°C . Under the same conditions, 0.3 to 0.5 vol % water was produced with FUETAP concrete containing 15 wt % simulated nonradioactive waste with or without 2.6 wt % NaNO_3 present. These results, which were obtained at ambient temperature to 130°C , showed no detectable difference in the amount of water produced.

The reaction rate of hydrogen and oxygen was cyclic in nature when the test gas contained $\geq 90\% \text{ H}_2$, 0.1 to 0.5% O_2 , and $<20 \text{ ppm H}_2\text{O}$. The rates were still 10 to 15 times greater in the presence of FUETAP concrete than with neat cement paste but were decreased by the presence of nitrate. The period and duration of the cycle were dependent on the water content of the concrete, the concentration of the oxygen in the gas mixture,

and possibly the oxidation state of the iron present as iron oxide in the simulated waste. This cyclic effect has also been demonstrated using iron oxide alone or with as little as 0.1 g of a partially oxidized iron wire. The cycling occurred at a higher temperature with the iron wire, 90 to 150°C, than was observed with concrete or iron oxide, 60 to 90°C, and was dependent on the pretreatment of the wire. Heating the wire to 200°C in a hydrogen atmosphere prior to testing resulted in sharper peaks and shorter cycling periods than were obtained with wire pretreated to 100°C.

Currently, plans are in progress to study radiolytic gas production in FUETAP concretes containing ^{244}Cm . The Safety Summary and the Quality Assurance Assessment were completed for these glove box tests, and work was initiated to develop the instrumentation required for the pressure measurements involved in the tests.

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3. STUDY OF CERAMIC AND CERMET WASTE FORMS (Activity No. AG 30 20 01 0; ONL-WH03; ON 1.3.1.2 AG)

W. S. Aaron, T. C. Quinby, E. H. Kobisk, D. K. Thomas

3.1 Summary

The current efforts in the cermet waste form development program, initiated in mid-1977, are centered on the improvement and refinement of processing techniques and the characterization of resulting cermets. High-level radioactive wastes from both defense and NFS sources are under consideration in this investigation. Significant improvements in processing techniques have been achieved, especially regarding the simplification of the process flowsheet and the development of a new densification technique. Quantitative evaluations of processing characteristics, such as radioisotope volatility losses and the effects of specific processing parameters on resulting cermet properties, are continuing. A leach testing program is currently being developed to avoid problems experienced in past cermet leach testing at ORNL and Battelle-Pacific Northwest Laboratory. Cermets have been formed from simulated wastes, radiotracer-containing simulated wastes, and three types of actual high-level wastes. A variety of examinations and analyses have been performed on these samples, and other tests are still in progress. At the current level of process and product development, limited engineering and economic feasibility studies have been initiated which will become more detailed and comprehensive as the program progresses.

3.2 Program Objectives

This program was initiated to develop and evaluate an alternative high-level radioactive waste solidification and fixation process which yields a cermet waste form.^{3.1} Background technology developed in the preparation of neutron dosimeter^{3.2} and various other special isotopic materials required only minor adaptations to be applicable to high-level

radioactive waste management.^{3.3} This cermet waste form is designed to incorporate most of the radioactive elements into micron-size particles of crystalline ceramic oxides, aluminosilicates, or titanates, all uniformly dispersed in an iron-nickel base metal matrix. The metal matrix which essentially micro-encapsulates the ceramic particles is composed of chemically reducible metals already in the waste (such as Fe, Ni, Cu, etc., and small amounts of reducible fission products) and those metals which are added to formulate a specific alloy composition. The metal matrix provides a high thermal conductivity, good corrosion resistance, and high mechanical strength to the product cermet. Efforts in this program include the development and quantification of the cermet process and a thorough characterization of the resulting cermet waste forms. These efforts are being carried out using a variety of simulated wastes, radiotracer-containing simulated wastes, and real high-level wastes from both defense and NFS sources.^{3.4}

3.3 Develop Continuous Calcination Technology

The precipitation and calcination steps in the cermet flowsheet have been successfully performed in one step using a rotary calciner. The calcined product was somewhat agglomerated, however, so a spray calciner was designed and is currently being fabricated. An ultrasonic spray nozzle has been incorporated in this calciner since it provides very efficient atomization of viscous fluids. It is expected that the calcine product from this spray calciner will be a fine, loose powder more amenable to subsequent densification than the rotary calciner product. The successful operation of the spray calciner, including an evaluation of processing parameter effects, is the key to continuous cermet processing. The development of continuous processing techniques has yielded the flowsheet shown in Fig. 3.1.

The spray calciner is being installed in a recently acquired trailer previously used as a chemical laboratory. In addition to being prepared for the calciner, the trailer will accommodate other developmental efforts in this program using simulated wastes.

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Figure 3.1 Process improvements simplify preparation of cermet high level waste forms.

3.4 Develop Cermet Densification Technique

The goal of this task is to minimize the void fraction and subsequent leachability of cermets through the selection and application of appropriate densification techniques. While hot pressing and isostatic pressing have been performed successfully, it is felt that cold compaction, by extrusion of the calcine, and sintering in a reducing atmosphere is the most promising densification method, especially where continuous processing is considered. Recent developments make this technique even more attractive. New cermet sintering techniques have been developed which significantly reduce the sintering temperatures and times necessary to achieve satisfactory densification. While further sintering parameter refinements are required, typical sintering conditions are currently 30 min at 1050°C as compared with the previous conditions of 24 hours at 1200°C or high temperature.

In conjunction with the development of high-temperature densification (sintering), various methods of cold-compaction of the calcine powder are being investigated. This work focuses primarily on the development of continuous extrusion techniques as would be appropriate for remote operation. Currently, conventional and unique extrusion die designs are being tested and evaluated.

3.5 Determine Process Volatility of Radioisotopes

As with any elevated temperature processing method, volatilization of radioisotopes may occur and the species must be identified for proper off-gas handling. Previous experiments using radiotracer-containing simulated wastes have shown very low losses of Cs, Ru, and Sr during all cermet processing steps.^{3.5} Experiments on the volatility of Tc are currently being initiated using simulated wastes containing ^{99}Tc with a $^{95\text{m}}\text{Tc}$ spike.

Volatility loss determinations made during the processing of real high-level wastes have supported the results obtained in the radiotracer

experiments. Unfortunately, due to a failure in the off-gas system to the processing hot cells, no volatility data were obtained during the processing of real NFS acid Thorex waste. The problem was corrected, however, and volatility data were obtained throughout the processing of Savannah River Plant (SRP) acid waste to form a cermet. Maximum total losses of 0.0033% and 0.2688% were determined for Ru and Cs, respectively. The greatest loss of Ru occurred during the calcination step, while the majority of the Cs losses occurred during the reduction/sintering process, as would be expected. Although not definitive, the lower volatility losses observed during the processing of SRP dried sludge to form a cermet, as compared to the acid waste experiment, support other indications that this cermet sample did not reach the appropriate sintering temperature due to misplacement in the furnace during remote loading.

3.6 Process Real High-Level Waste into Cermets

Evaluation of the NFS Acid Thorex cermet using optical and SEM metallography, thermal conductivity, calorimetry determinations, and physical dimension measurements has been completed. Due to equipment problems, no compositional analyses using the XES accessory on the in-cell SEM have been performed on this or any other cermets containing real waste. This pellet contained approximately 28 wt % waste and had a density of 6.27 g/cc, yielding a volume reduction factor of 4.5:1. The thermal conductivity of this pellet was found to range from 0.023 to 0.026 cal/sec·cm-°C over the 342 to 683°C temperature range. A total heat output of 0.157 watt or 0.024 watt/cc was determined by calorimetry. An SEM micrograph of this sample's structure is shown in Fig. 3.2. While the micro-encapsulation of the ceramic particles by the metal matrix was achieved, densification obtained in this first hot-cell run was not as great as desired nor as great as had been achieved in previous experiments.

The second hot-cell experiment involved the processing of fresh (<1 year out of the reactor) SRP acid waste into a cermet. This sample had a density of 6.44 g/cc. Because of the high Na content of this

Figure 3.2 SEM micrograph of the cermet formed from actual NFS acid Thorex waste.

waste and the high activity, it was decided to limit the waste loading in this first processing experiment to 2.1 wt %. A subsequent volume reduction factor of only 1.6:1 was observed which should not be considered a maximum because the limits on Na content and activity level in cermet have not yet been determined. The cermet sample had a decay heat output of 1.29 watts or 0.202 watt/cc and thermal conductivity values ranging from 0.0334 to 0.0466 cal/cm·sec-°C over a temperature range of 430 to 766°C. It has been calculated that a cermet bar 6 in. in diameter with this heat loading and thermal conductivity would possess a center-to-surface-temperature gradient of approximately 30°C. Optical metallography has been performed on this sample, and SEM metallography is in progress.

The last experiment in the series to form one cermet sample from each of three different real waste types, namely the conversion of SRP sludge to a cermet, was not entirely successful. Along with the previously mentioned volatility losses, the shrinkage during sintering and density of this sample were lower than expected, indicating incomplete reduction and/or sintering. The pressed pellet was probably misplaced into a cooler zone of the furnace during the remote loading operation. This conclusion was further supported by the inability to unload the furnace in the conventional manner. No detailed characterization of this sample has been performed.

Due to limited funding, further hot-cell experiments have been postponed. All equipment and the remainder of the three different wastes are being stored in-cell until this phase of the program is reinstituted. Leach tests on the previously formed cermet samples, additional processing runs, and further characterization of the actual waste cermets are planned at that time. At the request of DOE, based on our previous experience, arrangements were made for the sampling of NFS-treated waste supernate stored at West Valley, New York, and the transportation of this sample to ORNL for analysis. As of this date, the sample has been taken and is currently in transit to ORNL.

3.7 Determine Cermet Leach Rate and Compatibility

A series of leach tests, performed at PNL on simulated NFS Acid Thorex waste cermets containing cesium and ruthenium radiotracers, were recently completed and the samples returned to ORNL. Leachant solution analyses have been completed, and the samples are currently being sectioned and examined metallographically at ORNL. The six different leach tests performed at PNL on the cermet samples were as follows:

1. 72-hour Soxhlet test;
2. 31-hour, 25°C, pH 4 acetate buffer test;
3. 31-hour, 25°C, pH 9 ammonium hydroxide-HCl buffer test;
4. 146-day, long-term modified IAEA leach test at 25°C in distilled water with decreasing sampling frequencies;
5. 7-day, 350°C, 2500-psi distilled water autoclave test; and
6. 7-day, 350°C, 1550-psi saturated NaCl brine autoclave test.

Due to testing methods, galvanic coupling between cermet samples and metals in the test equipment was, unfortunately, present in five of the six PNL tests. The role of galvanic coupling in these particular tests has not been quantified; however, there is obvious evidence that it did occur. It should also be noted when evaluating these results that the leach rate expression of $\text{g/cm}^2 \cdot \text{day}$ is very dependent on the surface area of the sample. A geometric surface area was used to calculate cermet leach rates, but since the actual surface area is larger than the geometric area the leach rates listed in Table 3.1 are too high. Cermets are formed by densification of a powder precursor rather than a fusion process as used for glasses, for example. Therefore, their surface roughness is far greater than that of a fused waste form, yielding a higher active leaching surface area. The use of geometric surface areas is justifiable when comparing fully developed waste forms; however, the densification techniques for cermets have not yet been developed to the point where the present surface characteristics (porosity and surface roughness) are considered optimum. Therefore, these leach data should be evaluated with the factors of galvanic coupling and surface area estimates in mind.

Table 3.1. Results of PNL Leach Tests a, b, c

	(a) Soxhlet test, 72 hr	(b) pH 4, 31 hr	(c) pH 9, 31 hr
Wt. loss basis:			
% loss g/cm ² ·day	0.002 7.1 x 10 ⁻⁶	0.11 7.4 x 10 ⁻⁴	0.0024 1.6 x 10 ⁻⁵
Cs analysis basis:			
% Cs release g/cm ² ·day	0.024 7.18 x 10 ⁻⁵	0.072 4.75 x 10 ⁻⁴	0.015 2.43 x 10 ⁻⁵
Ru analysis basis:			
% Ru release g/cm ² ·day	N.D. ^a N.D. ^a	0.0016 1.02 x 10 ⁻⁵	0.0003 1.72 x 10 ⁻⁶

^aN.D. = no detected leaching.

The results of tests a, b, and c are shown in Table 3.1. Galvanic coupling was present in the Soxhlet test, but not in tests b and c. The leach rate for cesium was 7.18×10^{-5} g/cm²·day in this test. During identical testing of a PNL 78-68 solid glass core, an actual leach rate of 9×10^{-4} g/cm²·day was determined on a weight loss basis.^{3.6} Rates based on individual isotopic analyses were not available for the glass sample. Interpretation of the results from tests b and c is complicated by the actions of galvanic coupling between the cermet samples and the stainless steel mesh baskets which were used to suspend them in the leachant.

The long-term modified IAEA leach test (test d) was recently terminated after 146 days. Again, the cermet sample was suspended in the leachant by a Type 304L stainless steel mesh basket. During late stages of the test, a plastic basket was substituted for the stainless steel mesh. The leach rates determined in this test varied sporadically

in the 10^{-6} g/cm²·day order of magnitude, based on cesium analyses, throughout most of the test. At this time, because of the sporadic behavior and testing complications, no conclusions can be drawn from this test regarding cermet leach mechanisms.

Two hydrothermal leach tests were performed by PNL on cermet samples (tests e, f). Galvanic coupling was again a factor in these tests as the samples were held in Inconel 600 gaskets inside the Hastelloy C autoclaves. In the distilled water test, a weight gain of 0.0025% was observed while analyses of the clear leachant showed a total Cs release of 12% corresponding to a calculated leach rate of 1.5×10^{-2} g/cm²·day. The lustrous metallic surface of the cermet was transformed to a dull gray, matching the appearance of the autoclave surfaces. In the brine test, a weight gain of 2.82% was observed while analyses of the leachant indicated a 38% Cs release corresponding to a calculated leach rate of 3.9×10^{-2} g/cm²·day. A black adherent coating had developed on this sample. In both tests, the cermet samples appeared to maintain their physical integrity and no local attack or pitting was apparent from a visual examination of the surfaces.

The cermet samples from the PNL tests have recently been returned to ORNL, and metallographic examination of these samples is beginning. In initial examinations, the micrographs of sample cross sections in Figs. 3.3 and 3.4 indicate minimal attack of the samples in the long-term and the brine autoclave tests. In the long-term test, penetration depths of reaction layers did not exceed 5 microns and these reaction layers were only present under surface iron oxide layers which are assumed to have formed as a result of galvanic coupling with the stainless steel mesh basket used to support the cermet in the distilled water leachant. Qualitative analyses using XES indicate that a significant amount of Cs was not lost from aluminosilicate particles at, and just below, the surface. However, quantitative comparisons of the Cs content in these and interior particles have not been performed. In the brine autoclave test, no attack was detected metallographically beyond 100 microns below the surface and only local attack along apparent cracks was observed in the 100-micron-thick surface layer. Metallographic studies have not yet been initiated on the other four samples.

Fig. 3.3. SEM micrograph of cermet sample from test (d) showing surface deposit apparently resulting from galvanic coupling (30X:150X).

Fig. 3.4. SEM micrograph of cermet sample from test (f) showing minimal attack under hydrothermal conditions (30X:300X).

A leach testing program is now being formulated, taking into account difficulties previously experienced at ORNL and PNL, which should yield quantitative leach rate data on cermets at this stage of their development. These tests will be performed on cermets containing key radiotracers and on the cermets containing the real wastes.

3.8 Engineering Evaluation of Economic Feasibility

An estimate of metals usage and supply was prepared. Since a large portion of the cermets metal matrix is made up of iron and other metals already in the water, only small additions are required to formulate an iron-base alloy composition providing corrosion resistance, high thermal conductivity, and mechanical strength. Currently, large stockpiles of contaminated metals, unsuitable for commercial use, are held by the DOE. These stockpiles are estimated to include 17,000 tons of Ni, 30,000 tons of steel, and 1,700 tons of Cu with future inventory increases for these metals ranging from 500 to 7000 tons per year.^{3.7} It has been estimated that the processing of all SRP sludges existing in 1985 (31 tanks, 3.4 million gallons)^{3.8} in cermet form would require additions of from 160 to 810 tons of Ni and from 40 to 200 tons of Cu. This calculation assumes that the cermet metal matrix has a composition of 70% Fe, 20% Ni, 5% Cu, and 5% miscellaneous metals (such as Mo or Co, etc.). It can be seen that the amounts of metal additives required for the solidification of this large amount of waste are but a small portion of the current and projected contaminated metal inventories.

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