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OAK RIDGE WASTE MANAGEMENT PROGRAMS:

GEOLOGIC ISOLATION AND ACTINIDE PARTITIONING*

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INTRODUCTION

There are two waste management R&D programs of national significance that are being administered for ERDA by the Union Carbide Corporation -- Nuclear Division (UCC/ND). The National Waste Terminal Storage (NWTS) program is concerned with the development of geologic repositories for commercial nuclear fuel cycle wastes, and is organized as the Office of Waste Isolation within UCC/ND. The second program has the aim of establishing the technical feasibility and incentives for removing actinide elements from wastes ("partitioning") to reduce the long-term risk of terminal storage. The latter program is at Oak Ridge National Laboratory, and is only now getting under way. This paper summarizes the scope and objectives of these two programs and describes the principal elements and schedule for each.

GEOLOGIC ISOLATION PROGRAM

The National Waste Terminal Storage (NWTS) program has been initiated by the Energy Research and Development Administration as a part of a greatly expanded national effort in waste management. The objective of this program is to provide terminal storage facilities for commercial high-level and transuranic radioactive waste in deep geologic formations at multiple geographic locations. The design of these facilities will be based upon conventional techniques for handling radioactive materials and conventional underground excavation procedures, both modified as necessary for this application and the characteristics of the particular rock type and location. For each of the facilities, a major milestone will be the construction and operation of a pilot plant. These pilot plants will be primarily experimental facilities for the confirmation of all prior work, proof testing of waste handling equipment and techniques and demonstration of the suitability of waste disposal in the particular formation. They will receive and handle real radioactive wastes on a limited basis and will maintain all radioactive materials in a readily retrievable configuration. After an extended operational period as pilot plants, it would be possible to expand these facilities to full-scale waste repositories.

In order to provide a redundant waste capacity on a timely basis and to provide for accepting solidified commercial high-level wastes as they become available, important targets of the NWTS program are that the first two pilot plants be in operation simultaneously in 1985, followed by two additional similar facilities in 1987. In view of the severe constraints of this schedule, it is anticipated that the first two pilot plants will be established in salt formations. The data base and experience with natural salt deposits resulting from prior programs over a long period of time greatly exceeds that of other likely geologic repository formations.

The structure and content of the programs required to design and construct each of the pilot plants will be straightforward and similar in principle if not in detail to that required for any major nuclear project. However, the programs required to reach the initiation of a facility construction project are unique in many regards. For this reason, and because the bulk of the initial effort of the NWTS program will be expended on these preliminary studies, the remainder of this paper will be devoted to a description of these programs, a summary of their current status, and a discussion of specific plans for their development in the near future.

Conceptually, it is convenient to categorize the programs into three main activities:

1. The geological studies and investigations required to confirm the suitability of particular formations for waste disposal and to identify and certify specific sites.
2. In situ testing programs needed to define the response of the rock to waste disposal operations and to provide the specific data for facility design. Also included in this category are the design studies comparing alternative approaches and systems analysis required to establish optimum design criteria.
3. A broad range of technical support activities covering those studies and analyses which are relatively independent of rock type.

The interrelationships between these program activities and the way in which they are integrated to achieve the overall program objectives can be illustrated by examining the principal stages in the progression leading to a waste repository for a hypothetical and generalized case. The starting point is the technical judgment that a certain rock type in a particular geologic setting may be suitable for waste disposal based upon general geologic principles. The first step in the evaluation of the rock type is an in-depth review of the available information on the properties and characteristics of the formation. This review is regional or basin-wide in scope and emphasizes those features which are important for waste disposal. These preliminary studies result in the confirmation (or denial) of the suitability of the formation and produce supporting documentation that the original premise was valid. These preliminary studies also serve to identify specific areas (perhaps as large as 1000 square miles) qualifying for further investigation.

The next step involves much more detailed study of those promising areas including the development of new data from field geologic work, land surveys, geophysical surveys, geological exploration drilling and other special geological investigations. At the same time, the engineering properties of the formation are being examined by a series of in situ tests which are, in turn, supported by appropriate laboratory, analytical and theoretical studies. The first of these tests would be very limited in scope and intended primarily to provide a basis for more elaborate subsequent tests. These preliminary tests might, for example, consist of a few electrical heaters installed in the formation, or even in a mineralogically similar formation and could be at or near the ground surface.

Because these preliminary tests are "scouting" in nature, examining the gross response of the formation to the effects of waste (usually limited to the effects of heating), no attempt would be made to rigorously simulate either the geometry or the properties of real waste.

Depending upon the results of both the preliminary tests and the geological investigations, the next step would be field tests where an effort would be made to simulate actual waste disposal conditions while still limiting the scope of the experiments as much as possible. These field tests might be carried out in existing mines or other available excavations in the formation of interest or in similar formations, or perhaps they will be performed in near-surface excavations specially constructed for the purpose.

These field tests will be scheduled so that the results are available at about the same time the specific geological investigations of the study areas are nearing completion. Those geological investigations, together with the field tests, again serve to confirm (or deny) the suitability of the particular formation and provide the basis for identifying several much smaller locations (of perhaps 10-20 sq miles each) showing promise of containing potential sites for facilities. The next step then is the site-specific geological studies required to locate, and more importantly, to confirm the geological characteristics and properties of those sites. These investigations will involve extensive and concentrated exploratory drilling and testing of both core samples and in the hole.

At the same time, the next phase of in situ experiments would be undertaken. This phase is called "Vault Tests" and their objective is to simulate all aspects of an actual facility (including radioactivity) as closely as possible on a much reduced scale. These vault tests could be conducted in specially excavated portions of existing mines in the formation of interest, if available. In at least some of the formations, suitable existing excavation will not be available and it will be necessary to construct the entire experimental facility. In these cases the vault test may be located at a promising repository site but this would not be an essential requirement.

Following the identification and confirmation of a suitable site and the successful performance of a testing program to demonstrate the compatibility of waste disposal operations with the formation, a pilot plant as described above would be established. Present plans call for the first two of these pilot plants to be in operation by 1985 with the next two following in 1987 and subsequent pilot plants as needed at later dates. This schedule is necessary to assure that waste storage capacity is available at the time the first commercial high-level waste is delivered. This approach of establishing a number of facilities at a number of locations and in a number of different formations provides several advantages. The first of these is redundant storage capacity. Since the pilot plants are experimental facilities with all emplaced waste maintained in a readily retrievable condition, it is necessary to provide a reserve capacity should it ever be desired to exercise the retrieval option. A second advantage is that the possibility of utilizing a variety of geologic formations and rock types provides a further redundancy in that geologic storage can still be carried out even if a particular formation is disqualified for some fundamental reason. Finally, the establishment of a number of geologic terminal storage facilities at a number of locations permits the optimization of the entire waste management system including the minimization of shipping distances between reprocessing plants and storage facilities.

A large number of geologic formations and rock types are currently being investigated for their utility for waste disposal. The status of these programs is summarized below:

Natural Salt Deposits

Salt deposits were first identified as holding promise for use in waste disposal in 1955. Since that time, a large quantity of in situ testing, laboratory analysis and conceptual design development work has been carried out. Because of this previous experience and the availability of test results, it is anticipated that the first two pilot plants will be in salt formations. Based on preliminary reconnaissance surveys, the most promising regions for possible sites for these two plants are (1) the interior province of the Gulf Coast salt dome basin, especially in northeastern Louisiana, and (2) the bedded Salina formation in the Michigan basin portion of the Appalachian basin, specifically in the northeast quadrant of the lower penisula. In both cases, the preliminary reviews have been completed and the second stage geological studies are in progress.

These studies consist of geological exploration core drilling, seismic reflection geophysical surveys, special hydrology investigations and, for the domes, an intensive effort to evaluate the tectonic stability of a few selected structures. In addition, an in situ test is in preparation in an existing mine in a salt dome. This test is very limited in scope, involving only three electrically heated containers simulating high-level waste (radioactive sources will not be used), and is intended to validate the extrapolation of the large quantity of data from bedded salt experiments (primarily from the Project Salt Vault experiment at Lyons, Kansas) to dome salt deposits.

Other salt formations are also being studied at the preliminary review level, primarily as alternatives to the Michigan and dome salt deposits. These are the Permian Basin in southeastern Utah. It should also be recognized that the Delaware Basin portion of the Permian Basin in south-eastern New Mexico is also being investigated as a possible repository for ERDA generated radioactive wastes as a separate and independent program.

<u>Argillaceous Formations</u>

In order to provide the desired redundancy, it is intended that the second pair of pilot plants will be established in formations other than salt. In general, one of the more promising non-salt rock types appears to be the fine-grained sedimentary rocks containing large amounts of clay minerals - especially shales, mudstones and claystones. These rocks have many of the advantageous properties of salt, such as extremely low permeability that provides isolation from circulating ground water, and high plasticity. In addition, they usually exhibit a high ion exchange capacity that would retard radio-nuclides should they ever be dissolved in ground water. Several possible shale formations are being investigated on a preliminary review basis including the very wide-spread Pierre Formation of western North and South Dakota and Nebraska, and eastern Montana, Wyoming and Colorado; the several relatively small but contained Triassic shale basins along the east coast; certain clay formations of the Gulf Coast region and portions of the paleozoic shales of the Illinois Basin. This latter formation shows particular promise for waste disposal and is currently considered a target formation for an early pilot plant. However, it should be emphasized that whereas there has been a long history of testing in salt, very few and very limited testing has been completed in shale type rocks. There are a number of as yet unanswered questions concerning the behavior of argillaceous formations in the temperature and stress fields to which they will be subjected in waste disposal operations. These questions are particularly related to the structural stability of the particular formation in underground excavations and the changes in thermal, mechanical, chemical and mineralogical properties resulting from elevated temperatures. Most of these questions can be adequately answered only by in situ field testing at a reasonable scale. The first (and preliminary) test is currently in preparation at a near-surface site in the Conasauga shale formation in the Oak Ridge area. Although this formation is not currently of interest in itself for waste disposal, it is mineralogically and mechanically quite similar to other paleozoic shales, including those of the Illinois Basin, and therefore serves admirably for preliminary testing.

Crystalline and Other Rocks

Other rock types of interest include the flood basalts of the Columbia River Plateau; the Eleana Formation at the Nevada Test Site (which is a thick, highly indurated formation ranging from quartsitic shale to argillaceous quartzite); certain granitic intrusives, particularly at depths below the zone of circulating ground water, and; certain deeply buried, unjointed and unfractured limestones and other carbonates. The Columbus limestone in central Ohio appears to possess the necessary properties and is currently considered as a prospect for an early pilot plant.

ACTINIDE PARTITIONING PROGRAM

One possible way to manage high-level waste is to separate the actinides from the fission products, and thus reduce the duration of containment that is required for the residual fission-product-bearing wastes. Figure 1 shows what might be accomplished if these separations can be made from high-level waste by very high degrees of completion. The hazard index of waste is plotted against the age of the waste and the hazard indices of pitchblende and carnitite, both naturally

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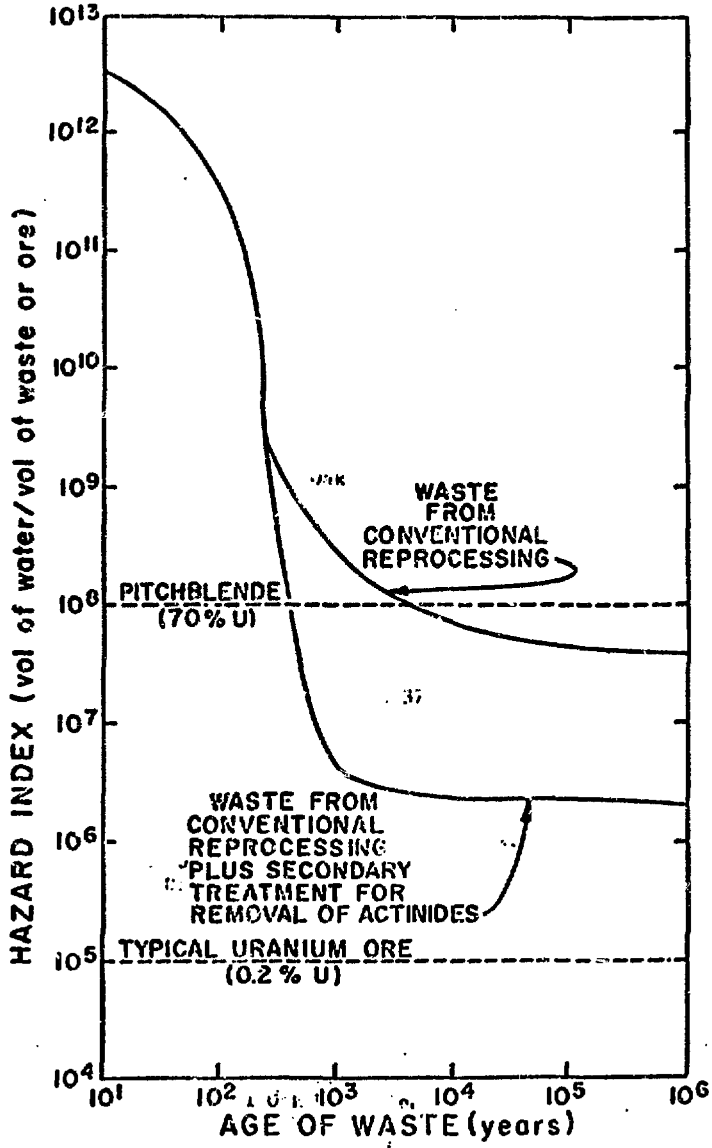

Fig. 1. Effect of age and method of treatment on the hazard index of high-level wastes from LWRs.

occurring radioactive substances, are shown as references. After 10^3 to 10^4 years, the hazard index* of conventional HLW drops and remains in the neighborhood of pitchblende. If one recovers greater than 99.99% of the Pu, 99.9% U, Am, Cm, and 95% of the Np in the spent fuel, the hazard index after 10^3 years is reduced by a factor of 20 to 50, and falls well within the range of widely distributed, naturally occurring materials. A thousand years' containment can be provided with high confidence by geologic storage; the argument becomes less convincing when confinement must be maintained for hundreds of thousands of years.

There are several prerequisites for this concept to become a realistic waste management option, however. First, exceptionally sharp separations of the six-or-more actinides from the fission products on a routine, day-to-day plant operating basis must be achieved without materially complicating the management of the remaining fission-product effluents. This means that all side streams from the processes must be recyclable, and that the final, processed fission-product waste must be of such a nature that it can be solidified and handled by techniques which are currently being developed. Second, while high-level wastes are always emphasized, it must be recognized that, if we are going this route, the actinides must be removed from all the wastes in which they appear. For

*The "hazard index" is defined as the volume of water required to dilute a unit volume of a radionuclide mixture to the levels defined in the Code of Federal Regulation (10 CFR 20) as the maximum permissible concentration for unrestricted use of water.

example, about 1% of the total Pu recycled is lost in the wastes from fuel preparation and fabrication, and with the cladding hulls. Finally, assuming that we have separated the actinides, we must be able to dispose of them. And within present or near-future technology this means burning them to fission products in power reactors. This can probably be done, but the implications of doing so on other parts of the fuel cycle (i.e., reactor operation, fabrication, and transportation) have not as yet been fully explored.

Although a modest amount of work has been done on both partitioning and transmutation,[1,2] it has been rather diffuse and uncoordinated. There remain large gaps in the information that many believe is needed for a convincing assessment of this concept. Consequently, ERDA has asked ORNL to initiate a broadly based program consisting of both experimental and computational tasks that is required to develop a meaningful cost-risk-benefit analysis of actinide partitioning and transmutation as a waste management concept. Several ERDA installations having specialized experience and experimental facilities will participate in the program. It is expected that at the end of the three-year program, reprocessing and refabrication flowsheets will exist which have been largely verified by experimental work; a cost-risk-benefit analysis will have been completed; and the scope and magnitude of the future development program needed to implement this concept will be defined. ERDA should then be in a position to judge whether or not partitioning and transmutation is a waste management alternative worthy of development through the demonstration stage.

Because of the recognized need for an early assessment of the partitioning-transmutation concept, the program will be limited to applied, rather than basic, research and development. Only those partitioning and transmutation options which presently show promise from an engineering viewpoint will be considered.

The program is divided into two group of tasks. The first group is comprised largely of experimental work concerned with separations; the second group is concerned with nonpartitioning aspects. These tasks are described below.

Development of Separation Flowsheets

The proposed future experimental program on flowsheet development is outlined in Table 1. The objective is to develop flowsheets, based on the firmest experimental evidence possible, of processes for removing the long-lived, biologically significant nuclides from all reprocessing and refabrication waste streams and convert them to forms suitable for burning in fission reactors. The studies are coordinated in such a manner as to minimize all residual losses and accumulations of all biologically significant elements recycled for burning, and to minimize the volumes of all secondary wastes generated. In order to achieve these goals it will be necessary to restrict the chemical additives which can be utilized in processing, and to produce only those secondary waste streams which can be recycled without an adverse effect on the overall plant operation.

Table 1. Development of separations flowsheets

(Funding allocation by task)

Task	Title and Site	$1000 FY-77	$1000 FY-78
A.	Recycle of waste streams (RF)	115	150
B.	U, Np, Pu recovery with TBP (ORNL)	270	300
C.	Actinide recovery from solids (ML)	70	100
	Actinide recovery from solids (ORNL)	0	100
D.	Am-Cm recovery (ORNL)	270	300
	Am-Cm recovery (SL)	50	50
	Am-Cm recovery (INEL)	125	180
E.	Long-lived isotope recovery (ANL)	160	175
F.	Actinide recovery from combustible waste (RF)	115	175
G.	Radiation effects (BNL)	50	50
H.	Flowsheet studies (ORNL)	70	70
	Task subtotals	1,295	1,650

An experimental approach to flowsheet evaluation has been chosen because the complex chemistry of electrolytic solutions, typical of streams found in fuel reprocessing, cannot be adequately modeled at this time. Moreover, the concept requires that all biologically significant species be recovered with losses to all waste streams which are substantially less than have been demonstrated in the past. There is no adequate, theoretical basis for estimating these losses. Consequently, flowsheet analysis and evaluation which is not based on experimental observations is largely speculative, and in the context of this study cannot be construed to demonstrate technical feasibility.

Task A. - Studies will be initiated at Rocky Flats (RF) on the recovery of actinides from secondary waste streams likely to be produced in fuel fabrication and reprocessing; the residual effluent concentrations will define the purity of recycled acid and water streams. Both cation exchange and mixed ion exchange beds will be used, and the application of reverse osmosis will be examined.

Task B. - Both cold and full-radiation-level mixer-settled tests on the extraction of U, Np, and Pu will be done at ORNL. These studies will be made to demonstrate high recovery as well as the concept of recycling low- and intermediate-level liquid waste streams will be acceptably low.

Task C. - The leaching of actinides from HEPA filters and other secondary solid wastes such as failed equipment without the addition of chemicals which preclude subsequent recycle of the decontamination solutions to a Purex plant will be studied at Mound Laboratory (ML). The leaching of actinides from dissolver and high-level liquid waste solids, and their fusion for actinide recovery will be studied at ORNL during FY-1978.

Tasks D and E. - A multisite experimental program for the recovery and purification of Am and Cm will be initiated. Oxalate precipitation, cation exchange chromatography, and solvent extraction (Talspeak Process) will be tested at ORNL; separation by elution on an inorganic ion exchanger medium such as a titanate or zirconate will be studied at Sandia Laboratories (SL); recovery by bidentate extraction from the HLLW will be considered at the Idaho Nuclear Engineering Laboratories; and the feasibility of recovering macro-amounts of actinides and Tc from HLLW using phosphorous- and amine-based extractants and extraction chromatography will be studied at Argonne National Laboratory.

Task F. - Rocky Flats will investigate the feasibility of a recovery process which involves solubilizing incinerated wastes, if possible, or leaching the actinides from the ash.

Task G. - A literature search summarizing existing knowledge on the effects of intense radiation fields in the degradation of all process chemicals, and secondary reactions caused by radiation, will be undertaken by Brookhaven National Laboratory (BNL). Some radiation damage studies with these materials may be performed as a part of this task.

Task H. - Flowsheet evaluation using computer modeling is the last task of this group, and this will serve as a focal point for the analysis and orientation of all the experiemental work.

Final partitioning flowsheets for reprocessing and refabrication will be selected by the end of the second year, and we expect that most of the experimental work will cease. The mass rates and compositions of all streams will be defined along with energy balances. The expected losses for all waste streams, as defined by the experimental work and flowsheet analyses, will be specified.

Nonpartitioning Aspects

The objectives of the nonpartitioning tasks (Table 2) are to (1) determine the feasibility and in-reactor effects of transmuting the long-lived nuclides in fission reactors, (2) determine the impact of the increased handling of the long-lived nuclides on fuel cycle operations other than reprocessing, (3) perform a risk-benefit analysis on the entire partitioning-transmutation concept, (4) update the ORIGEN computer code to reflect the most current neutron cross-sections, decay data, and reactor models available. A fifth task concerns the coordination and analysis of the total program.

Task I. - The transmutation calculations will involve both LWRs and LMFBRs, since the LWRs will constitute the principal available transmutation neutron source until well into the next century and the LMFBRs are probably superior in an overall sense as transmutation reactors. Savannah River Laboratory will look at transmutation strategies for LWRs and the Neutron Physics Division of ORNL will concentrate on LMFBRs. The transmutation program because the in-reactor behavior of the recycled, long-lived

Table 2. Nonpartitioning aspects

(Funding allocation by task)

Task	Title and Site	$1000 FY-77	$1000 FY-78
I.	In-reactor transmutation (ORNL)	70	80
	In-reactor transmutation (SRL)	70	80
J.	Fuel cycle impact studies (ORNL)	70	160
K.	Risk-benefit analysis (PNL)	120	240
L.	ORIGEN updating (ORNL)	140	40
M.	Coordination and analysis (ORNL)	105	170
	Subtotals	575	770
	Totals	1,870	2,420

nuclides in various configurations will define the form (target assemblies, target rods, or homogeneously dispersed in the fuel) in which the long-lived nuclides are recycled. The form in which the recycled, long-lived nuclides are irradiated can have a major impact on the rest of the nuclear fuel cycle. For instance, concentrating the actinides causes greater neutron shielding requirements during fabrication, transportation, reprocessing, and refueling; a reduced transmutation rate due to self-shielding effects; the presence of a concentrated neutron "sink" in the reactor; and a smaller amount of highly neutron-active material to be fabricated. As a result of complex fuel cycle interactions such as these, the in-reactor transmutation studies are inextricably intertwined with the other parts of the partitioning program and should, in fact, be conducted as part of the partitioning program.

Task J. - The investigation of the partitioning-transmutation fuel cycle impacts is important since the major impacts of partitioning and transmutation, excluding the reprocessing plant, are expected to be felt in the transportation and fabrication sectors because of the increased neutron activity of the long-lived nuclides. These impacts must be analyzed in detail for realistic long-lived nuclide recycle modes if the risk-benefit calculation, which uses the results of the impact analysis, is to be meaningful.

Task K. - The risk-benefit analysis that will be done at Pacific Northwest Laboratories utilizes the results of all previous tasks to estimate the additional risk incurred as a result of increased handling of the long-lived nuclides, and the benefits which would accrue as a result of the biologically significant, long-lived nuclide content of radioactive wastes being significantly reduced. Work in 1977 and 1978 will consist mainly of developing the methodology and applying it to the conventional case of managing fuel cycle wastes without partitioning.

Tasks L and M. - The last two tasks, updating the ORIGEN computer code and coordinating the program do not require great elaboration. ORIGEN is a widely-used tool for many types of fuel cycle analysis, design, and planning work, and we are already deeply involved in updating and broadening its nuclear data base. This work will continue and the code will be maintained in the best condition possible for use on this and other programs.

Expected Results in FY 1979

The third year of the program will be spent principally in completing the analyses, including an analysis of costs, and documenting the results of the study. The sectors of the nuclear fuel cycle which will be analyzed for increased risk are fabrication, fresh-fuel transport, transmutation reactor safety, spent fuel shipping, and the reprocessing-partitioning-waste solidification facility. If all goes as planned, the program should supply a quantitative estimate of the incremental differences in total costs, risks, and benefits to be derived from implementing this fuel cycle waste management concept, as opposed to isolating the bulk waste mixtures in geologic formations as is now intended. It should also provide a description and schedule of the future research, development, and demonstration program that would be needed to reduce the concept to practice.

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