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HIGH-LEVEL WASTE MANAGEMENT RESEARCH AND DEVELOPMENT PROGRAM
AT OAK RIDGE NATIONAL LABORATORY*

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1. INTRODUCTION

Since the mid-1950's, Oak Ridge National Laboratory has been actively engaged in the development and evaluation of waste management methods for the expanding nuclear power industry. Much of this work was a normal outgrowth of our earlier involvement with fuel reprocessing studies, since it is in reprocessing that many of the most formidable waste management problems arise. Until recently, the principal developmental approach to the management of high-level wastes was to convert them to stable solid forms suitable for permanent disposal in a bedded salt formation. However, within the last year or so the AEC has broadened its program in this area to include the evaluation of alternative approaches. Our participation in this new work consists principally of feasibility studies of separating the actinide elements in the wastes from the fission products to reduce the long-term risk in disposal, and evaluations of other geologic formations than bedded salt for their suitability as disposal media.

Today, I would like to review the status of our work in both of these areas. However, first I would like to summarize very briefly our most recent projections of the high-level wastes to be generated by the nuclear power industry over the remainder of this century.

2. WASTE PROJECTIONS

Although all forecasts are inexact and often controversial to some extent, they nevertheless can serve as the best guide to future conditions, and thus

have become for most of us a useful tool for planning and evaluation purposes. Recently, we completed projections of all the wastes that would arise from the nuclear fuel cycle over the remainder of this century, but I would like to present here only our forecast of high-level wastes.

The projections were based on a recent AEC forecast of nuclear power growth in the U.S. [WASH-1139 (72)], and on fuel mass-flow data developed for ^{235}U - and Pu-enriched LWRs, an HTGR, and two IMFBRs using a systems analysis computer code. Nuclear characteristics of the fuels and wastes were computed using the ORIGEN code.

The projected installed nuclear electric capacity is shown in SLIDE 1. The contribution of each of the three types of reactors is shown here through the year 2000. The total installed nuclear capacity is expected to rise from 5 GW at the end of 1970 to 13⁴ GW in 1980, and to 1200 GW in the year 2000. In this projection, the greatest proportion of the capacity is contributed by LWRs, with HTGRs coming on-stream in 1977. The IMFBRs go on-stream in 1987, overtake the installed HTGR capacity in 1995, and almost overtake the LWRs by the end of the century.

SLIDE 2 presents the spent fuel reprocessing requirements and high-level waste conditions for this projected nuclear power economy. The metric tons of spent fuels to be reprocessed annually rise from 2400 in 1980 to 17,600 in the year 2000, and we estimate that this function will be carried out in only 8 or 10 plants distributed throughout the country. The estimates of annual generation of high-level liquid wastes assume 100 to 300 gal of waste per ton of fuel reprocessed, and the accumulated volume of liquid and solid wastes assume the wastes are stored as a liquid for 5 years before solidification.

PROJECTED INSTALLED NUCLEAR CAPACITY IN THE
U.S.

PROJECTED FUEL REPROCESSING LOAD AND HIGH-LEVEL WASTE GENERATION

	FISCAL YEAR		
	1980	1990	2006
FUEL REPROCESSED ANNUALLY, TONS	2400	8400	17,600
WASTE GENERATED ANNUALLY, 10 ⁶ GAL	0.24	0.97	2.3
ACCUMULATED PROPERTIES			
VOLUME OF LIQUID, 10 ⁶ GAL ^a	0.92	3.8	10.0
VOLUME OF SOLID, CU FT	1000	61,000	270,000
TOTAL RADIOACTIVITY, MCI	10,000	50,000	150,000
TOTAL HEAT-RATE, MW	49	250	670
TOTAL INHALATION HAZARD ^b	1.8×10^{20}	3.9×10^{21}	7.3×10^{21}
TOTAL INGESTION HAZARD ^b	2.4×10^{15}	1.5×10^{16}	5.0×10^{16}

^a WASTE STORED AS LIQUID FOR FIVE YEARS BEFORE SOLIDIFICATION.

^b CUBIC METERS OF WATER REQUIRED FOR DILUTION TO RCG.

The inhalation and ingestion hazards as used here are defined as the cubic meters of air or water that would be required to dilute all the radioactive constituents to the concentration specified in the RCG (10 CFR 20) as being suitable for unrestricted use. The hazards given for the year 2000 compare with about 10^{19} and 10^{18} m³, reported to be the volumes of the earth's atmosphere and surface waters, respectively. Such comparisons as this can be very misleading, however, unless it is recognized that it is impracticable to disperse globally significant quantities of deeply buried wastes. Processes such as insolubilization, sedimentation, ion-exchange, and filtration all limit dispersibility and provide for self-purification of the atmosphere and oceans.

SLIDES 3, 4, and 5 present nuclear characteristics of all the wastes accumulated through the year 2000 as a function of time following that date. We have assumed that the actinide elements in the waste consist of 0.5% of the uranium and plutonium originally in the spent fuels, and all other heavy elements formed by transmutation of U and Pu in the reactors. The fission products predominate in terms of the thermal generation and ingestion hazard for the first several centuries, but after a thousand years decay, the actinides alone constitute the hazard. On the basis of inhalation hazard, the actinides predominate even initially. This very fact has given rise to the question of why not remove the actinides from the fission products in the wastes and thus reduce the containment requirement from a million-or-so years to one of only a thousand years, or less.

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KILOWATTS OF THERMAL POWER IN HIGH-LEVEL WASTES TO BE
ACCUMULATED THROUGH THE YEAR 2000

	YEAR 2000	TIME FOLLOWING 2000 (YEARS)		
		1000	100,000	1,000,000
FISSION PRODUCTS	614,000	4.9	2.9	0.18
ACTINIDES	50,700	937	31	19
TOTAL	665,000	942	34	19

SLIDE 3

INGESTION HAZARD OF HIGH-LEVEL WASTES TO BE ACCUMULATED
THROUGH THE YEAR 2000^a

	YEAR 2000	TIME FOLLOWING 2000 (YEARS)		
		1000	100,000	1,000,000
FISSION PRODUCTS	5.0×10^{16}	1.7×10^{10}	1.3×10^{10}	2.3×10^9
ACTINIDES	2.2×10^{14}	1.8×10^{13}	2.3×10^{12}	6.8×10^{11}
TOTAL	5.0×10^{16}	1.8×10^{13}	2.3×10^{12}	6.8×10^{11}

^a CUBIC METERS OF WATER REQUIRED FOR DILUTION TO RCG
(COLUMN 2 OF TABLE II, 10CFR20).

INHALATION HAZARD OF HIGH-LEVEL WASTES TO BE ACCUMULATED
THROUGH THE YEAR 2000^a

	YEAR 2000	TIME FOLLOWING 2000 (YEARS)		
		1000	100,000	1,000,000
FISSION PRODUCTS	6.7×10^{20}	1.7×10^{15}	1.3×10^{15}	2.0×10^{14}
ACTINIDES	6.6×10^{21}	2.2×10^{20}	7.4×10^{18}	4.1×10^{18}
TOTAL	7.3×10^{21}	2.2×10^{20}	7.4×10^{18}	4.1×10^{18}

^a CUBIC METERS OF AIR REQUIRED FOR DILUTION TO RCG
(COLUMN 1 OF TABLE II, 10CFR20).

3. WASTE PARTITIONING

SLIDE 6 illustrates the possible merits of such separations by comparing the hazard index of the wastes from conventional processing of fuel from a typical LWR with the wastes resulting from a postulated secondary treatment wherein the five principal actinides (U through Cm in the periodic table) are removed by factors of 10^3 to 10^5 . It is also assumed that 99.9% of the iodine has been separated during reprocessing. The hazard index is defined as the volume of water that could be contaminated to the RCG-allowable value by one volume of waste or other radioactive material. For reference, this slide also shows the hazard index associated with the mineral pitchblende and with a uranium ore containing 0.2% U, typical of the large deposits that occur on the Colorado Plateau. Note that the hazard index of the waste from conventional processing decreases rapidly over the first thousand years (due to the decay of ^{90}Sr and ^{137}Cs) but remains as great or greater than pitchblende for more than a million years. The waste resulting from secondary treatment, however, falls within the range of naturally occurring radioactive materials after only several hundred years, a time span for which we can reliably extrapolate the effects of geologic, climatic, and other natural phenomena. This type of waste then could surely be buried in geologic formations with negligible risk to future generations. One way of disposing of the separated actinides and iodine would be to recycle them with fresh fuel to power reactors, and our calculations show that even after sustained recycle of these materials, the wastes, while having a slightly higher index would still lie well within the range of naturally occurring radioactive species.

EFFECT OF AGE AND METHOD OF TREATMENT ON THE HAZARD INDEX OF HIGH-LEVEL WASTES FROM LWR'S

SLIDE 7 presents the percentage removal of each actinide element in the wastes from different reactor fuels that would be required to reduce the hazard index of the wastes to less than 5% of that of pitchblende after storage for about 1000 years. (Further reduction of actinides would not be warranted unless one also removed the very long-lived fission products: ^{99}Tc , ^{93}Zr , ^{135}Cs , and additional ^{129}I .) As a general statement, we can say that we must remove 99.99% of the Pu, 99.9% of the U, Am, and Cm, 95% of the Np, and 95% of the Th and Pa if we are processing HTGR fuels. Separations processes currently used to recover actinides for commercial or scientific uses have given Pu and U recoveries of about 99.5%, and Np, Am, and Cm recoveries in the range of 90 to 99%.

It is, therefore, apparent that a comprehensive development program will be required to reduce this proposed waste management concept to practice. This program must be designed (SLIDE 8) to (1) solve problems that are obvious from previous experience, (2) increase the removal of actinides to the desired levels, (3) determine the most desirable method to integrate the needed process cycles into an overall system, (4) choose chemical processes and reagents that minimize high-level waste treatment problems, and (5) determine the composition of all waste streams that will be generated and ways to recycle these streams.

We have begun our investigations by limiting our considerations to wastes from LWR fuels because this is the only type for which experience on large-scale reprocessing exists. Concentrations of key constituents and required recovery factors are shown in SLIDE 9. We have constructed

PERCENTAGE REMOVALS REQUIRED TO REDUCE HAZARD INDEX
OF AGED WASTES TO <5% OF THAT OF PITCHBLEND^a

ACTINIDE	PWR-U	PWR-Pu	LMFBR	HTGR
Th				95.5
Pa				95.0
U	99.9	99.9	99.9	99.99
Np	95.0	95.0	70.0	95.0
Pu	99.95	99.99	99.99	99.9
Am & Cm	99.0	99.9	99.9	95.0

^a MINERAL CONTAINING 60% U.

SLIDE 7

OBJECTIVES OF ACTINIDE SEPARATIONS PROGRAM

1. SOLVE PROBLEMS OBVIOUS FROM PREVIOUS EXPERIENCE.
 2. INCREASE REMOVAL OF ACTINIDES TO DESIRED LEVELS.
 3. DETERMINE HOW TO INTEGRATE NEEDED PROCESS CYCLES INTO AN OVERALL SYSTEM.
 4. CHOOSE PROCESSES AND REAGENTS THAT MINIMIZE HIGH-LEVEL WASTE TREATMENT PROBLEMS.
 5. DETERMINE HOW TO RECYCLE ALL GENERATED WASTE STREAMS.
-

SLIDE 8

**COMPOSITION OF HIGH-LEVEL WASTE FROM REPROCESSING
LWR FUEL AND ACTINIDE RECOVERY FACTORS REQUIRED**

CONSTITUENT	CONCENTRATION (g/LITER)	RECOVERY FACTOR REQUIRED
U	0.93	≥ 5
Np	0.15	≥ 20
Pu	0.009	≥ 50
Am	0.03	≥ 1000
Cm	0.007	≥ 1000
TOTAL F.P.	5.60	
R.E.'s	2.03	

**BASIS: 33,000 MWd EXPOSURE; 5175 LITERS WASTE PER TON OF
FUEL; 0.5% LOSS OF U AND Pu TO WASTE.**

SLIDE 9

a variety of flowsheets based on previously established separations processes, considering it to be of paramount importance that waste streams generated by processes for recovery of the actinides from the wastes be suitable for recycle to previous stages or for combination and management with the residual fission-product waste. As of this time, only limited laboratory experiments with synthetic solutions of fission products have been done. Work with tracer levels of actinides will get under way shortly.

Our present conceptual flowsheets are based on the approach outlined in SLIDE 10. Uranium, Np, and Pu would first be recovered to the desired degrees using successive TBP extraction stages. The problems to be resolved here are concerned with (1) keeping Np in the extractable ^{+4 or} ~~+5 or~~ +6 valence states, and (2) the effect of solvent degradation products and other impurities on Pu recovery and on later separations.

Next, the waste would be stored for at least 5 years to reduce radiation damage to process reagents, solvents, or ion exchange resins to be used in subsequent separations. The problem here is that not only are large inventories of waste in liquid form accumulated, but also that Pu grows back into the waste during storage and may again require removal.

Following storage, Am and Cm are then separated using either the Talspeak or CEC process. The Talspeak process is based on solvent extraction with an organic phosphoric acid from a complexed carboxylic acid solution, while the CEC process uses cation exchange chromatography. Both processes are capable of separating Am and Cm from the lanthanides in the absence of other fission products. The problem here, then, is ^{to} prepare a suitable

PRESENT CONCEPTUAL FLOWSHEET APPROACH

1. RECOVERY OF U, Np, AND Pu TO DESIRED DEGREE USING SUCCESSIVE TBP EXTRACTION STAGES.
 2. STORAGE OF WASTES FOR ABOUT 5 YEARS.
 3. SEPARATION OF AMERICIUM AND CURIUM USING EITHER THE TALSPEAK OR CEC PROCESS.
-

SLIDE 10

feed for Talspeak or CEC by first separating Am, Cm, and the rare earths from the other fission products. We are considering two solvent extraction processes for this purpose: one using undiluted TBP, the other using di(2-ethylhexyl)phosphoric acid.

As I indicated earlier, there is much work yet to be done before even technical feasibility can be established. Once this is accomplished, we envision that another 10 to 15 years may be required for process development and demonstration of this waste management approach on a plant scale.

4. GEOLOGIC DISPOSAL EVALUATIONS

Following the unsuccessful attempt to establish a demonstrational waste repository near Lyons, Kansas, in 1970-1971, the AEC asked us to look elsewhere for a site where a pilot-plant repository might be established to confirm the suitability of bedded salt as a repository for high-level waste. With the help of the USGS and others, an area in southeast New Mexico was chosen as one worthy of intensive investigation (SLIDE 11).

Our current investigative program is outlined in SLIDE 12. We are drilling a 4000-ft-deep exploratory hole in S.E. New Mexico in which hydrological tests will be made and geophysical logs will be taken. A core will be taken for minerological studies and physical properties measurements. A local microseismic study of the area is under way by a consultant at New Mexico Tech. A study of the oil and gas potential of the area is being made by a consulting firm in Dallas. We are planning to carry out non-radioactive tests in an existing potash mine in New Mexico to obtain

0 10 20 30 MILES

LOCATION MAP

CURRENT INVESTIGATIONS OF BEDDED SALT

1. EXPLORATORY BOREHOLE IN S.E. NEW MEXICO.
 2. SEISMIC STUDY OF S.E. NEW MEXICO.
 3. STUDY OF OIL AND GAS POTENTIAL IN S.E. NEW MEXICO.
 4. NONRADIOACTIVE IN-SITU TESTS IN A S.E. NEW MEXICO MINE.
 5. BOREHOLE DISSOLUTIONING STUDIES.
 6. BOREHOLE PLUGGING STUDIES.
-

SLIDE 12

information on permeability, heat transfer, brine migration, and rock mechanics. Studies designed to clarify the mechanism of dissolution of salt in and around boreholes have been undertaken at two universities and by one private consultant. Methods and materials for use in permanently plugging boreholes are being studied at two universities. The borehole plugging problem is not unique to salt, of course, but must be faced if disposal is made into any type of geologic formation.

Thus far, all of our work has yielded most encouraging results, and we feel as confident as ever that bedded salt is completely suitable for the disposal of solidified high-level wastes. Furthermore, our relationship both with state officials and private citizens in New Mexico has been very good thus far. However, the Commission has made a recent decision to avoid a commitment to any type of geologic disposal in the immediate future, and as a consequence, we have been asked to broaden our program to include an evaluation of other promising geologic formations for storage or disposal of not only high-level, but of other types of wastes as well. The formations of greatest interest are salt anticlines and piercement domes, shales, granite, and limestone. We are only now reorienting our program and have not as yet formulated a detailed program plan to achieve the desired objectives. Nevertheless, this clearly represents an intention on the part of the AEC to develop a number of viable options for the possible future geologic disposal of many of the wastes that will be generated over the next several decades.