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ORIGIN, MAGNITUDE, AND TREATMENT OF RADIOACTIVE WASTES*

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MASTER

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ORIGIN, MAGNITUDE, AND TREATMENT OF RADIOACTIVE WASTES*

1. Introduction — Radioactive wastes in solid, liquid, and gaseous forms are generated wherever radioactive materials are handled. Unlike conventional industrial wastes, most of them are amenable (within presently available technology) to treatment that can destroy their toxicity. They become innocuous only through natural decay, and many of the isotopes of concern decay so slowly that they must be isolated from the environment for hundreds of thousands of years. The primary objective of nuclear waste management is to protect man and his environment from these materials by providing containment by means that are available within present or near-future technology.

   In the first of two talks on waste management, I would like today to define the problem by reviewing the origin and nature of the wastes, and the methods of treatment that are in use or that soon can be made available. Tomorrow, I will address the problem of their ultimate isolation, or isolation.

2. Origin of Wastes — Although the safe handling and disposition of radioactive waste effluents is an important concern wherever radioactive materials are present, it is within the various operations of the nuclear fuel cycle that the problem assumes its most formidable proportions.

   2.1 Schematic Depiction of the Fuel Cycle — DWG 68-12735R3.

   2.2 Radioactive Wastes from the Fuel Cycle — DWG 69-83R2.

   Wastes arise from each step of the fuel cycle, but most of the radioactivity is associated with fuel reprocessing and, to a lesser extent, with reactor operation.

   2.2.1 Wastes from mining, milling, refining, and fuel fabrication contain mainly isotopes from uranium and plutonium decay

   a. Mining — ^{222}Rn is diluted with air
   b. Milling — Tailings neutralized, sent to ponds for sedimentation, solar evaporation, or seepage; ^{226}Ra, ^{222}Rn, and ^{210}Pb are the most important isotopes
   c. Refining (conversion) — Contains mainly residual ^{226}Ra and is evaporated and stored
   d. Fuel fabrication — Solids from UO_2 fabrication are buried; liquids are clarified by sedimentation and/or filtration and released

   2.2.2 Wastes from reactor operation — Radioisotopes originate in the primary coolant and appear in wastes as by-products of purification. They arise from activation of corrosion products, chemical additives, and the coolant (H_2O) itself; from tramp U; from leakers.

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a. Characteristics — mostly very short half-lives
   Gaseous: only 8 isotopes after 30 min; only ^{85}Kr after 60 d, and the total activity then is only 10^{-6} that at 30 m.
   Liquid: over 90% of activity removed by simple filtration; f.p.'s normally much less significant than corrosion products; 3H derived from fission, n, reactions and B.
   Solids: come from treatment (filters, I-X resins) and many miscellaneous sources.

b. Operational Experience — Discharges have been typically less than 10% of limits on annual basis; ^{3}H less than 1%. New waste systems designed to even greater containment using noble gas recovery units and internal recycle of liquids. Future wastes from LMFBR's and HTGR's await development of those reactors, but "near zero" releases should be attainable.

2.2.3 Wastes from fuel reprocessing

a. Spent fuel — This is the source of virtually all of the radionuclides that are of concern. If the decision is made not to reprocess spent fuel, then it becomes a waste in the true definition of the word, so let's look at that first.

   Physical characteristics — DWG 78-529.

   Radioactivity — DWG 78-447. Both PWR and BWR assemblies contain more than a megacurie of activity 1 year after discharge, and in the neighborhood of 10^5 curies after 10-yr decay.

   Thermal power — DWG 78-458. The thermal power decreases by more than a factor of 10 between 1 and 10 years after reactor discharge.

   Concentrations of isotopes — DWG 69-13184R1. These values are for a metric ton of spent fuel. There is also about one curie of ^{14}C per ton of fuel from each of these reactors.

b. Diagram of origin of wastes — DWG 65-915. In addition to those shown here, will have iodine and tritium wastes, plus a very wide assortment of contaminated solid refuse.

c. High-level liquid wastes — DWG 70-6315. High-level wastes pose the greatest problem. They are HNO_3 solutions of about 400 isotopes of about 40 fission product elements and about 20 isotopes of about 6 heavy elements, and additionally contain varying concentrations of process reagents and corrosion products. These are the most significant radioactive constituents.

3. Waste Projections

3.1 Fuel Cycle Wastes Projected for the Year 2000 — DWG 78-636. This is an overestimate in that the basis is for an installed nuclear capacity of about 600 GW in 2000. We expect now something in the range of 200 to 400 GW, but the numbers should be internally consistent in a relative sense, and they do assume some Pu recycle in LWRs.

3.2 Fuel Cycle Wastes from 1000 MW-Years of Electricity — DWG 78-634. Pu recycle increases the volume of low-level TRU wastes but reduces ore requirements (and tailings) by ~40%.

3.3 Accumulated high-level wastes.

Megacuries of fission products in HLW — DWG 73-3963R1.
Megacuries of actinides — DWG 73-3964R1.
Kilowatts of thermal power — DWG 73-3965R1.
Ingestion toxicity — DWG 73-3966R1.
Inhalation toxicity — DWG 73-3967R1.

The preceeding series of slides raises the question of why not separate the fission products from actinides and simplify isolation requirements.

4. Waste Treatment

4.1 Options for Management of HLW — DWG 72-10392A. Three principal options are available.

4.2 Separations (Partitioning) and Transmutation — DWG 72-11707.

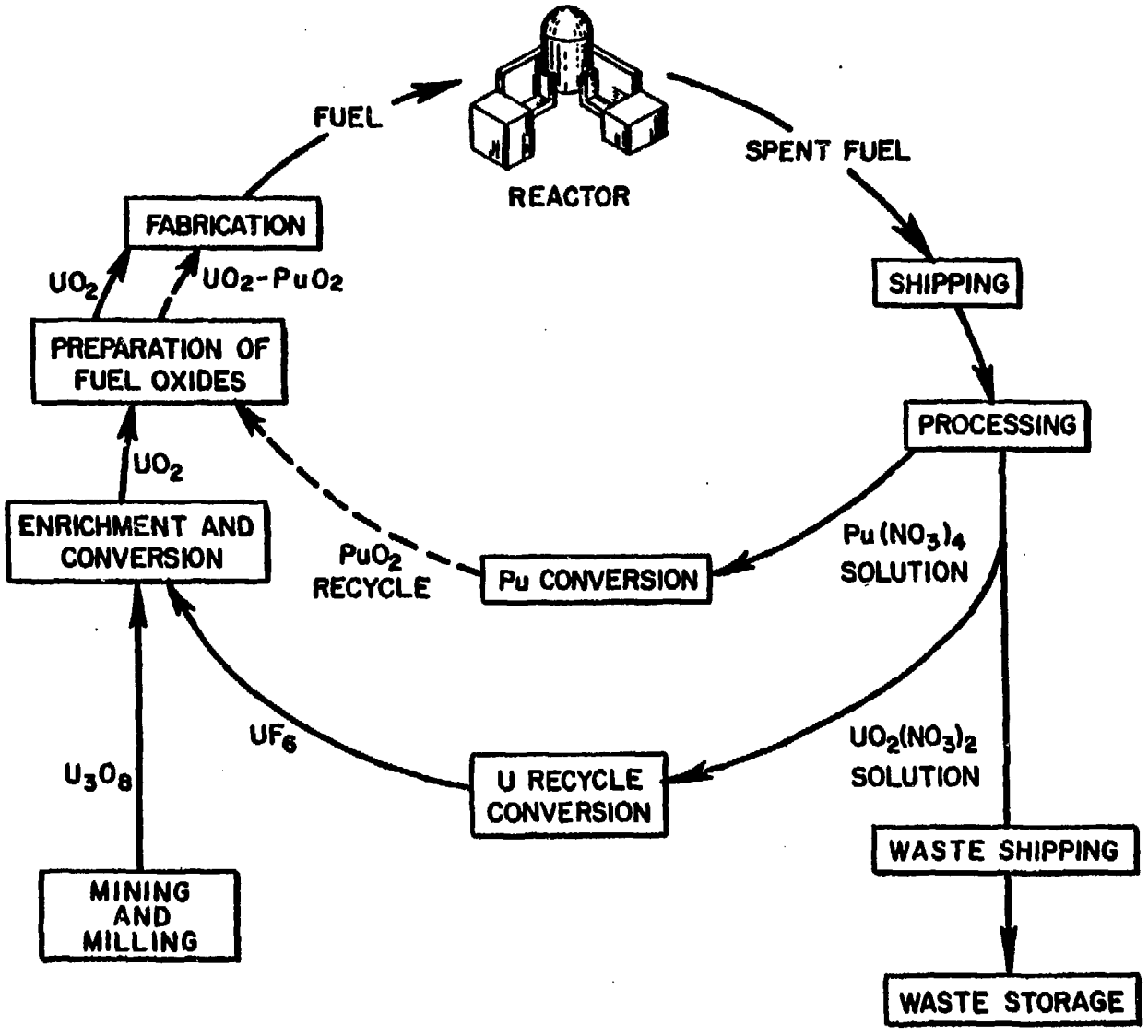
4.2.1 Requirements for actinide separations — DWG 75-5339.

4.2.2 Summary of transmutation studies — DWG 76-910. Study in final stages of completion indicates technical feasibility, but lack of incentives for development in view of costs and only nominal reduction of long-term risk.

4.3 Solidification. Many solid waste forms have been proposed and several carried to advanced stages of development and testing — DWG 79-1660. Glass has received most attention in U.S. and Europe and it does appear to be a satisfactory form; however, it may not be the very best, or ultimate form.

4.3.1 Photo of an aluminoborosilicate glass — PHOTO 61999. Processes have been developed both in this country and Europe to produce products of this type, and sizable amounts have been made on both prototype and commercial basis from actual high-level wastes. Furthermore, the glasses have been shown to be thermally and radiation-resistant at acceptable and achievable storage conditions.

NUCLEAR FUEL CYCLE FOR LIGHT WATER REACTORS



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ORNL DWG 69-83 R2

RADIOACTIVE WASTES FROM THE FUEL CYCLE

		Types of Wastes and Principal Constituents	Approximate Radioactivity Level (Ci/ton U)
Mining and Milling	Gaseous:	^{222}Rn, ^{218}Po, ^{214}Bi, ^{214}Po	10^{-4} - 10^{-3}
	Liquid, Solid:	U, ^{226}Ra, ^{230}Th, ^{210}Pb	0.5 - 1
Refining	Liquid:	^{238}U, ^{234}Th, ^{234}Pa, ^{226}Ra	10^{-4} - 10^{-3}
Fuel Fabrication	Liquid, Solid:	U, Pu, Th	10^{-4} - 10^{-3}
Reactor Operation	Gaseous:	^{13}N, ^{41}A, ^{89}Kr, ^{87}Kr, ^{138}Xe, ^{135}Xe	10 - 100[a]
	Liquid, Solid:	^{58}Co, ^{60}Co, ^{59}Fe, ^{51}Cr, ^{3}H	50 - 100[a]
Chemical Processing	Gaseous:	^{85}Kr, ^{133}Xe, ^{131}I, ^{129}I, ^{3}H	7000[b]
	Liquid, Solid:	Fission Products, Pu, Am, Cm	6,000,000[b]

[a] At time of waste discharge or shipment based on fuel exposure of 20,000 Mwd/ton of U.

[b] Waste from fuel at 20,000 Mwd/ton, 120 days cooled.

ORNL DWG 78-529

PHYSICAL CHARACTERISTICS OF TYPICAL UNIRRADIATED LWR FUEL ASSEMBLIES

	BWR	PWR
OVERALL ASSEMBLY LENGTH, M	4.47	4.06
CROSS SECTION, CM	13.9 x 13.9	21.4 x 21.4
ACTIVE FUEL HEIGHT, M	3.76	3.66
FUEL PIN O.D., CM	1.25	0.95
FUEL PIN ARRAY	8 x 8	17 x 17
ASSEMBLY TOTAL WEIGHT, KG	276	658
URANIUM/ASSEMBLY, KG	183	461
ZIRCALOY/ASSEMBLY, KG	57[A]	108[B]
HARDWARE/ASSEMBLY, KG	9.8[C]	26[D]
TOTAL METAL/ASSEMBLY, KG	68	135

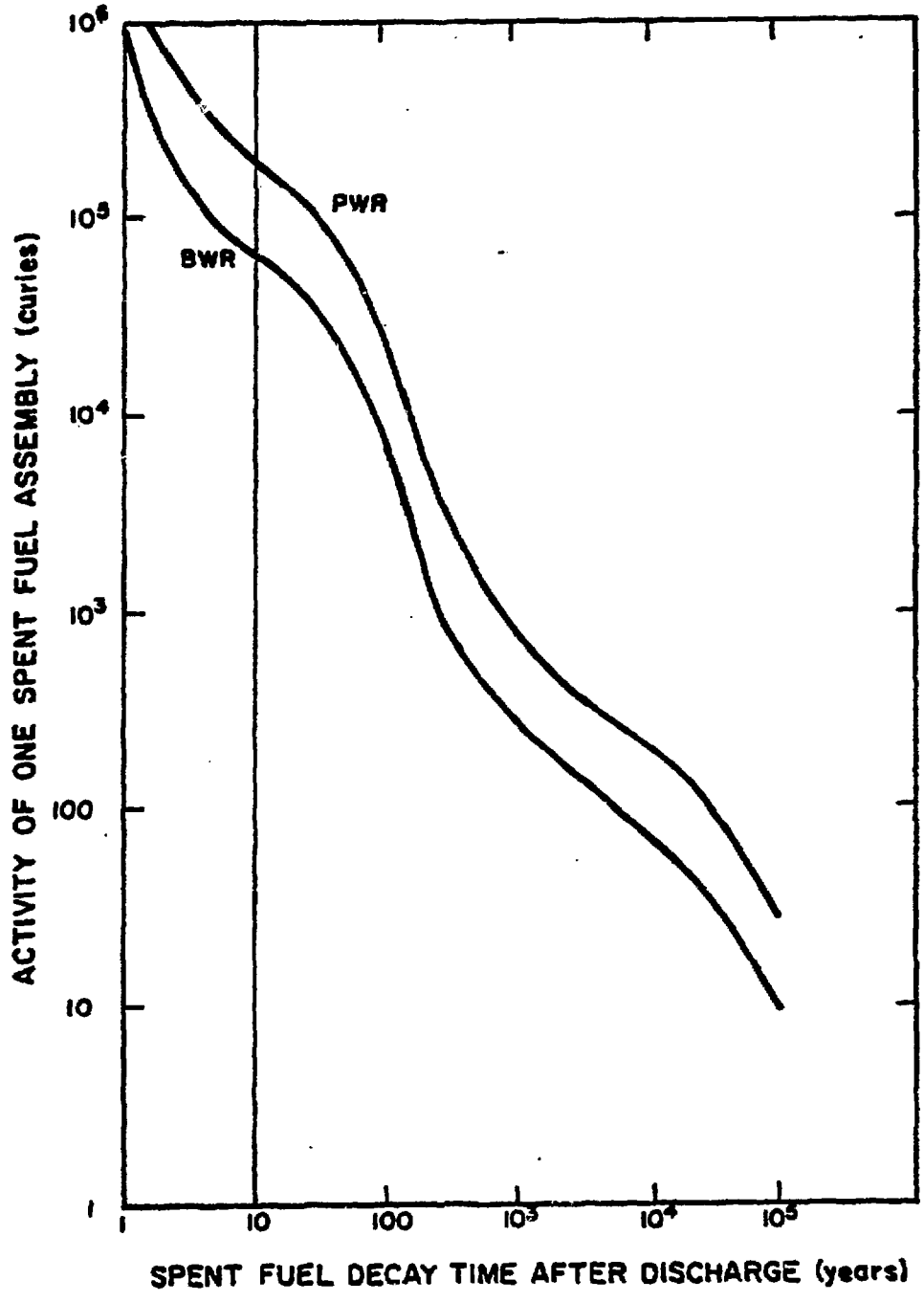
[A] INCLUDES ZIRCALOY FUEL-ELEMENT SPACERS.

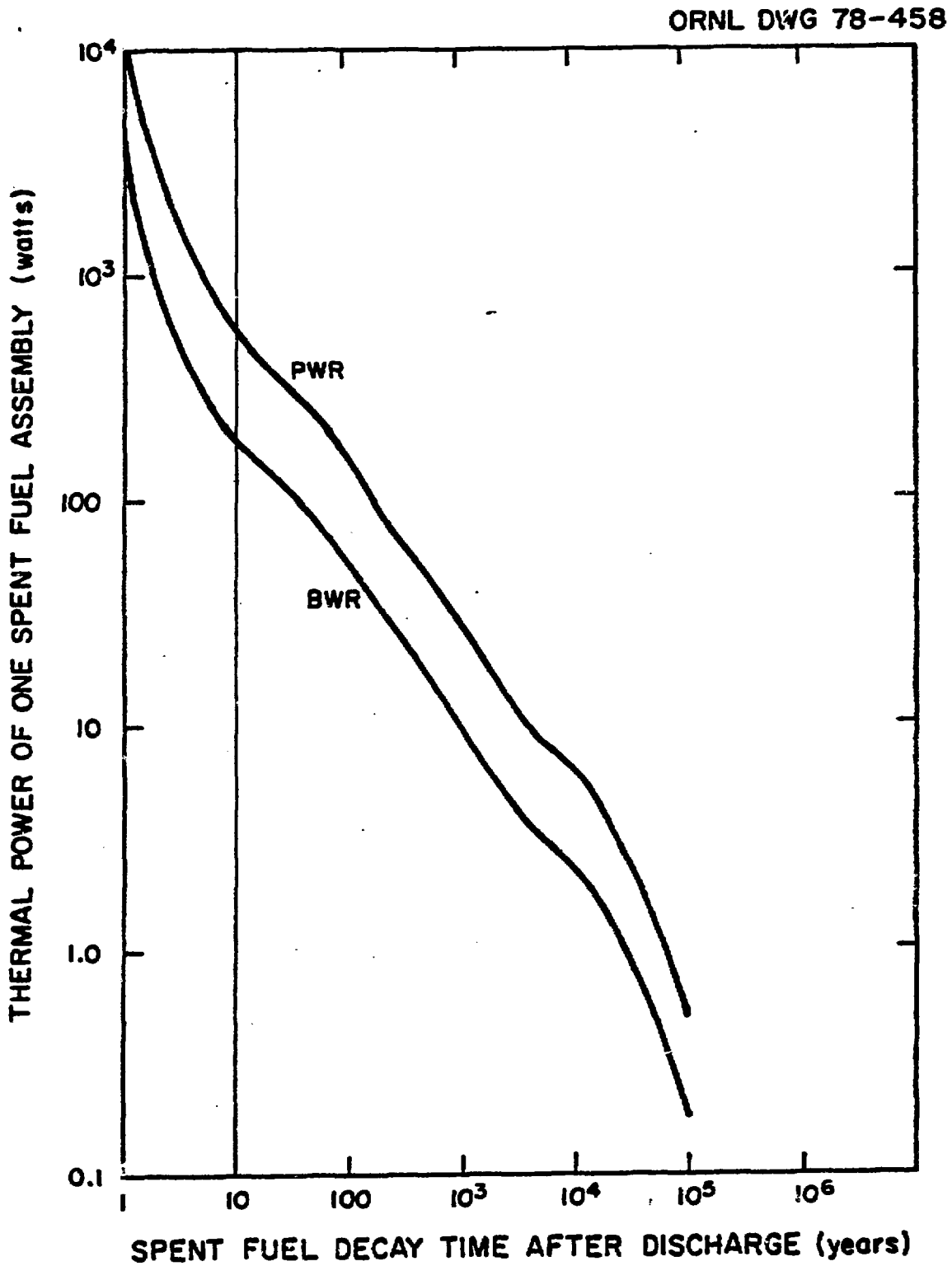
[B] INCLUDES ZIRCALOY CONTROL-ROD GUIDE THIMBLES.

[C] INCLUDES STAINLESS STEEL TIE-PLATES AND INCONEL SPRINGS.

[D] INCLUDES STAINLESS STEEL NOZZLES AND INCONEL-718 GRIDS.

ORNL DWG 78-447





THERMAL POWER OF SPENT BWR AND PWR FUEL ASSEMBLIES

ORNL DWG 69-13184 RI

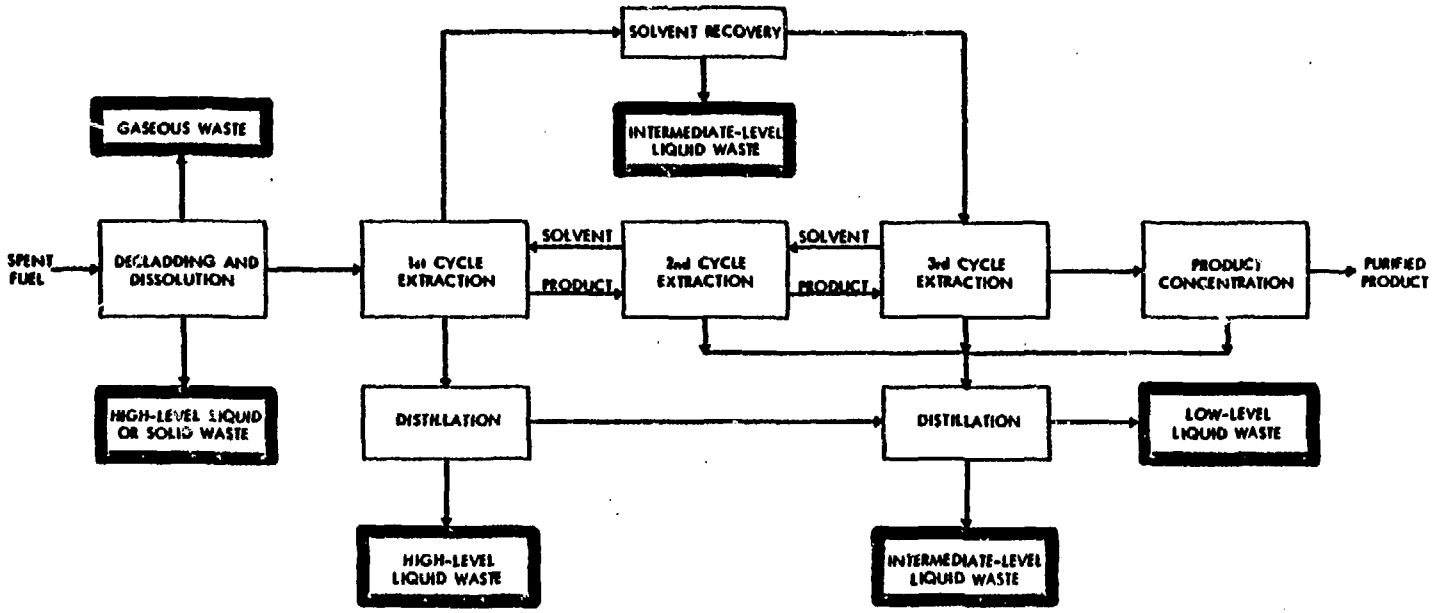
CONCENTRATIONS OF RADIOACTIVE MATERIALS IN SPENT REACTOR FUELS

SPECIES	MEGACURIES PER METRIC TON OF FUEL			
	LWR[a]		LMFBR[b]	
	DISCHARGE	150 DAYS	DISCHARGE	60 DAYS
TRITIUM	0.00070	0.00068	0.00094	0.00093
NOBLE GAS (^{85}Kr, ^{133}Xe)	11	0.011	25	0.013
IODINE (^{131}I, ^{129}I)	12	0.0000022	20	0.011
SEMIVOLATILES (Ru, Te, Cs, Se, Tc)	26	0.85	53	2.7
NONVOLATILE FP's (Sr, Zr, Rare Earths)	88	3.5	180	11
PLUTONIUM (^{238}Pu, ^{241}Pu)	0.46	0.12	1.1	0.62
Am-Cm (^{241}Am, ^{242}Cm, ^{244}Cm)	0.21	0.022	0.27	0.061

[a] LWR BURNUP = 33,000 Mwd/METRIC TON AT 30 Mw/METRIC TON.

[b] LMFBR CORE + BLANKET BURNUP = 33,000 Mwd/METRIC TON AT 58 Mw/METRIC TON.

ORNL DWG 65-915



Origin of Fuel Processing Wastes.

ORNL DWG 70-6315

SIGNIFICANT RADIOACTIVE CONSTITUENTS IN AQUEOUS WASTE FROM FUEL REPROCESSING

NUCLIDE	HALF-LIFE (YEARS)	CONCENTRATION[a] (μCi/ml)	10 CFR 20 LIMITS (μCi/ml)	YEARS TO DECAY TO MPC
^{3}H	12.3	4.6×10^{3}	3×10^{-3}	175
^{90}Sr	28.9	5.1×10^{5}	3×10^{-7}	1,200
^{129}I	1.6×10^{7}	0.254	6×10^{-8}	4×10^{8}
^{137}Cs	30	7.1×10^{5}	2×10^{-5}	1,000
^{244}Cm	18.1	1.7×10^{4}	7×10^{-6}	600
^{239}Pu	24,400	11.1	5×10^{-6}	500,000

[a] FUEL BURNUP OF 33,000 Mwd/TON AND 330 GAL WASTE PER TON.

ORNL DWG 78-636

FUEL CYCLE WASTES PROJECTED FOR THE YEAR 2000

CATEGORY OF WASTE	ANNUAL GENERATION VOLUME (10^3 m^3)	ANNUAL GENERATION ACTIVITY (MCi)	TOTAL ACCUMULATED VOLUME (10^3 m^3)	TOTAL ACCUMULATED ACTIVITY (MCi)	TOTAL ACCUMULATED POWER (MW)
SPENT FUEL ASSEMBLIES	3.4	40,800	40.1	86,400	340
TRANSURANIUM WASTES					
HIGH-LEVEL SOLIDIFIED	0.67	40,500	8.0	85,400	336
CLADDING HULLS	0.56	280	6.7	980	4.1
INT.-LEVEL SOLID	2.9	2.8	28	12.4	0.05
LOW-LEVEL SOLID	17	11	142	77	0.10
NON-TRU WASTES					
NOBLE GASES	0.02	59	0.22	470	0.7
IODINE	0.009	0.0002	0.1	0.003	1.6×10^{-6}
CARBON-14	-	0.004	-	0.05	-
LWR TRITIUM	53	0.3	660	2.1	7.0×10^{-5}
FP TRITIUM	0.07	3.5	0.8	29	1.0×10^{-3}
LOW-LEVEL SOLID	390	0.8	5500	5.7	-
ORE TAILINGS	15,000	0.18	224,000	2.5	0.06

ORNL DWG 78-634

FUEL CYCLE WASTES FROM THE PRODUCTION OF 1000 MW-YEARS OF ELECTRICITY[a]

CATEGORY OF WASTE	VOLUME (m^3)	ACTIVITY[b] (MCi)	POWER[b] (kW)	NUMBER SHIPMENTS
SPENT FUEL ASSEMBLIES	14	172	970	11
TRANSURANIUM WASTES				
HIGH-LEVEL SOLIDIFIED	3.1	170	960	2
CLADDING HULLS	2.7	2.0	10	2
INT.-LEVEL SOLID	140	0.012	0.057	66
LOW-LEVEL SOLID	480	0.05	0.04	17
NON-TRU WASTES				
NOBLE GASES	0.1	0.25	0.37	0.4
IODINE	0.05	1×10^{-6}	-	0.2
CARBON-14	-	2×10^{-5}	-	-
LWR TRITIUM	140	6×10^{-4}	-	9
FP TRITIUM	0.35	0.02	7×10^{-4}	1
LOW-LEVEL SOLID	2400	0.002	0.007	180
ORE TAILINGS	42,000	5×10^{-4}	0.011	-

[a]MIXED (U-Pu) OXIDE-FUELED LWRs.

[b]ACTIVITY AND THERMAL POWER AT TIME OF WASTE GENERATION.

ORNL DWG 73-3963R1

MEGACURIES OF FISSION PRODUCTS TO BE ACCUMULATED IN HIGH-LEVEL WASTES THROUGH THE YEAR 2000[A]

	YEAR 2000	TIME FOLLOWING 2000 (YEARS)		
		1,000	100,000	1,000,000
TOTAL FISSION PRODUCTS	77,000	2.4	1.7	0.35
^{90}Sr	6,600[B,C]	–	–	–
^{93}Zr	0.20	0.20	0.19	0.12[B,C]
^{99}Tc	1.6	1.6[B,C]	1.2[B,C]	0.06
^{137}Cs	10,000	–	–	–

[A]INSTALLED NUCLEAR CAPACITIES ARE 70, 285, AND 625 GW IN CALENDAR YEARS 1980, 1990, AND 2000. WASTE VOLUME IN THE YEAR 2000 WILL BE ABOUT 20 MILLION GAL OF LIQUID, OR 380,000 FT^3 IF SOLIDIFIED.

[B]CONTROLLING INHALATION HAZARD AT INDICATED TIME.

[C]CONTROLLING INGESTION HAZARD AT INDICATED TIME.

ORNL DWG 73-3964R1

MEGACURIES OF ACTINIDES TO BE ACCUMULATED IN HIGH-LEVEL WASTES THROUGH THE YEAR 2000[A]

	YEAR 2000	TIME FOLLOWING 2000 (YEARS)		
		1,000	100,000	1,000,000
TOTAL ACTINIDES	1,260	29	0.55	0.55
^{226}Ra	–	–	0.0058[C]	0.0012
^{229}Th	–	–	0.02	0.05[B,C]
^{239}Pu	0.24	0.39	0.17[B]	–
^{240}Pu	1.0	2.9	0.0001	–
^{241}Am	64	14[B,C]	–	–
^{243}Am	6.0	5.5	0.0007	–
^{244}Cm	820[B,C]	–	–	–

[A]ASSUMES LOSS TO WASTE OF 0.5% OF Pu, U, AND Th IN FUELS, AND ALL OF THE OTHER ACTINIDE ELEMENTS FORMED DURING IRRADIATION.

[B]CONTROLLING INHALATION HAZARD AT INDICATED TIME.

[C]CONTROLLING INGESTION HAZARD AT INDICATED TIME.

ORNL DWG 73-3965R1

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

	YEAR 2000	TIME FOLLOWING 2000 (YEARS)		
		1,000	100,000	1,000,000
FISSION PRODUCTS	330,000	2.7	1.6	0.096
ACTINIDES	41,000	780	14	13
TOTAL	371,000	780	16	13

ORNL DWG 73-3966R1

INGESTION TOXICITY OF HIGH-LEVEL WASTES TO BE ACCUMULATED THROUGH THE YEAR 2000[A]

	YEAR 2000	TIME FOLLOWING 2000 (YEARS)		
		1,000	100,000	1,000,000
FISSION PRODUCTS	2.5×10^{16}	9.4×10^{9}	7.0×10^{9}	1.2×10^{3}
ACTINIDES	1.5×10^{14}	5.6×10^{12}	4.2×10^{11}	3.1×10^{11}
TOTAL	2.5×10^{16}	5.6×10^{12}	4.3×10^{11}	3.1×10^{11}

[A]CUBIC METERS OF WATER REQUIRED FOR DILUTION TO RCG (COLUMN 2 OF TABLE II, 10CFR20).

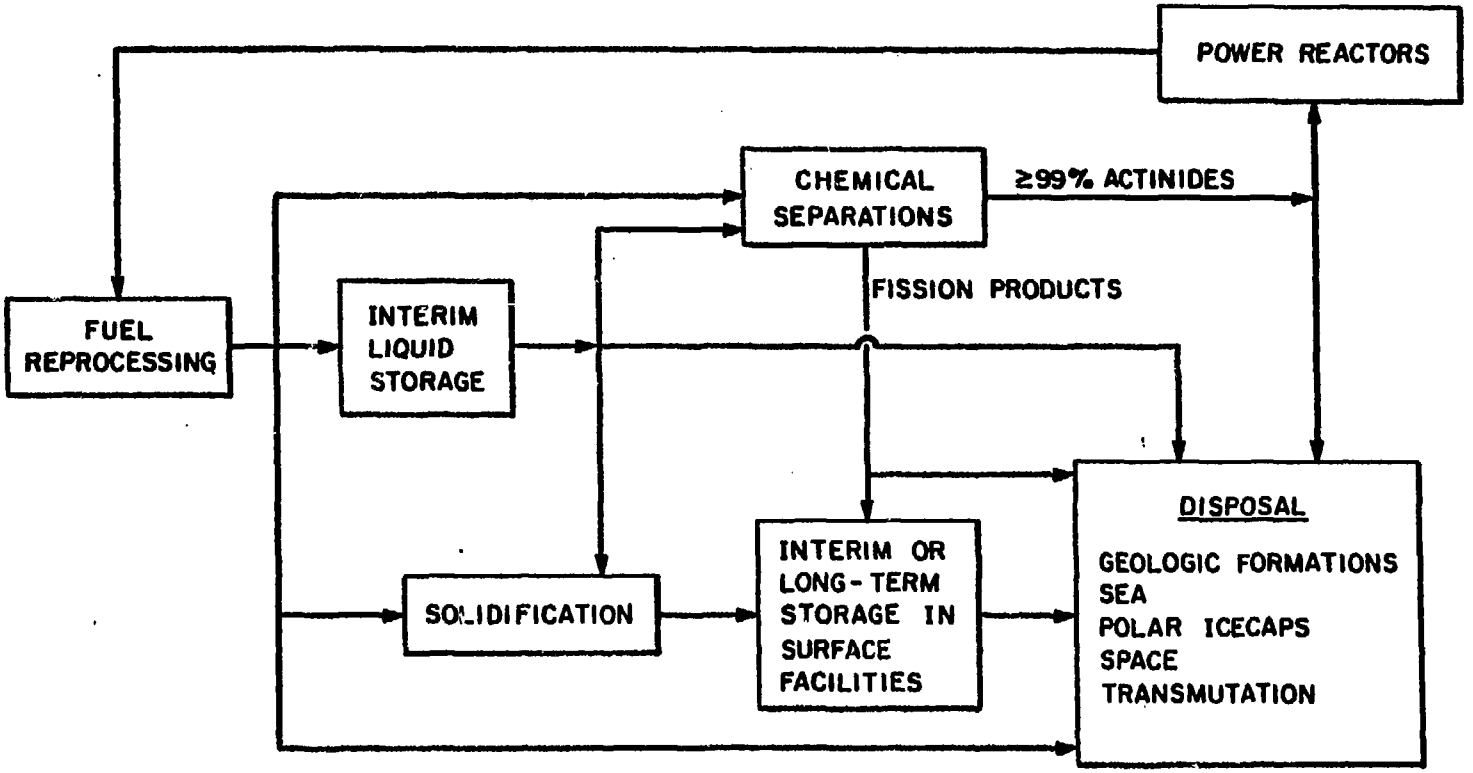
ORNL DWG 73-3967R1

INHALATION TOXICITY OF HIGH-LEVEL WASTES TO BE ACCUMULATED THROUGH THE YEAR 2000[A]

	YEAR 2000	TIME FOLLOWING 2000 (YEARS)		
		1,000	100,000	1,000,000
FISSION PRODUCTS	3.4×10^{20}	9.4×10^{14}	6.9×10^{14}	1.0×10^{14}
ACTINIDES	3.5×10^{21}	1.6×10^{20}	4.7×10^{18}	3.0×10^{18}
TOTAL	3.8×10^{21}	1.6×10^{20}	4.7×10^{18}	3.0×10^{18}

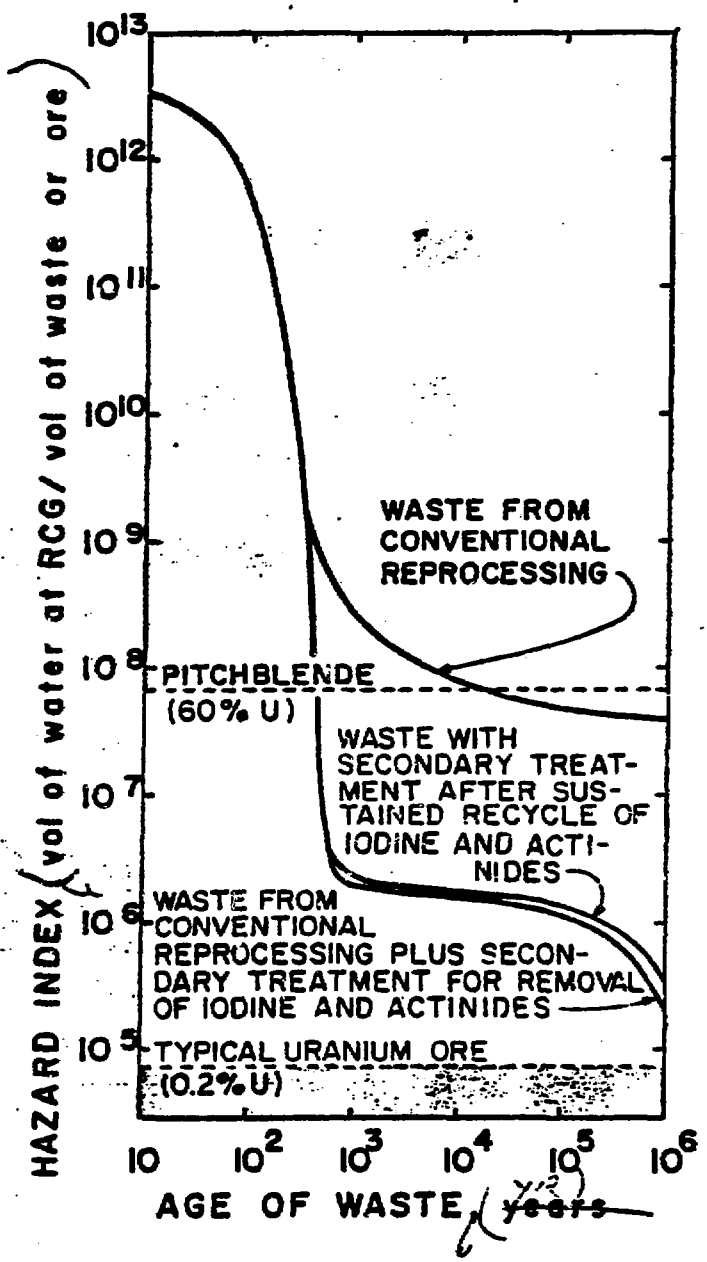
[A]CUBIC METERS OF AIR REQUIRED FOR DILUTION TO RCG (COLUMN 1 OF TABLE II, 10CFR20).

ORNL-DWG 72-10392A



Options for Management of High-Level Wastes.

ORNL DWG. 72-11707



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

ORNL DWG. 75-5339

REQUIREMENTS FOR ACCEPTABLE ACTINIDE SEPARATIONS CONCEPT

1. SEPARATIONS MUST BE MADE WITHOUT COMPLICATING HIGH-LEVEL WASTE SOLIDIFICATION, SHIPMENT, AND DISPOSAL.

2. SEPARATIONS MUST BE MADE FROM ALL ACTINIDE WASTES (~ 1% Pu LOST IN FUEL PREPARATION, FABRICATION AND CLADDING).

3. MUST BE ABLE TO BURN THE SEPARATED ACTINIDES WITHOUT UNDULY COMPLICATING OTHER FUEL CYCLE OPERATIONS.

ORNL DWG 76-910

SUMMARY OF TRANSMUTATION STUDIES TO DATE

1. ACTINIDE FISSION RATES ARE TYPICALLY 5 TO 7%/YEAR IN BOTH LWRs AND LMFBRs.

2. REACTOR OPERATING CHARACTERISTICS ARE NOT GREATLY IMPAIRED: K_∞ REDUCED ~1%; MASS ~2% OF FUEL.

3. NEUTRON ACTIVITY OF FUELS IS INCREASED SEVERAL ORDERS OF MAGNITUDE ABOVE NORMAL LEVELS.

ORNL-DWG 79-1660

ALTERNATIVE SOLID WASTE FORMS

CALCINE

CONCRETE

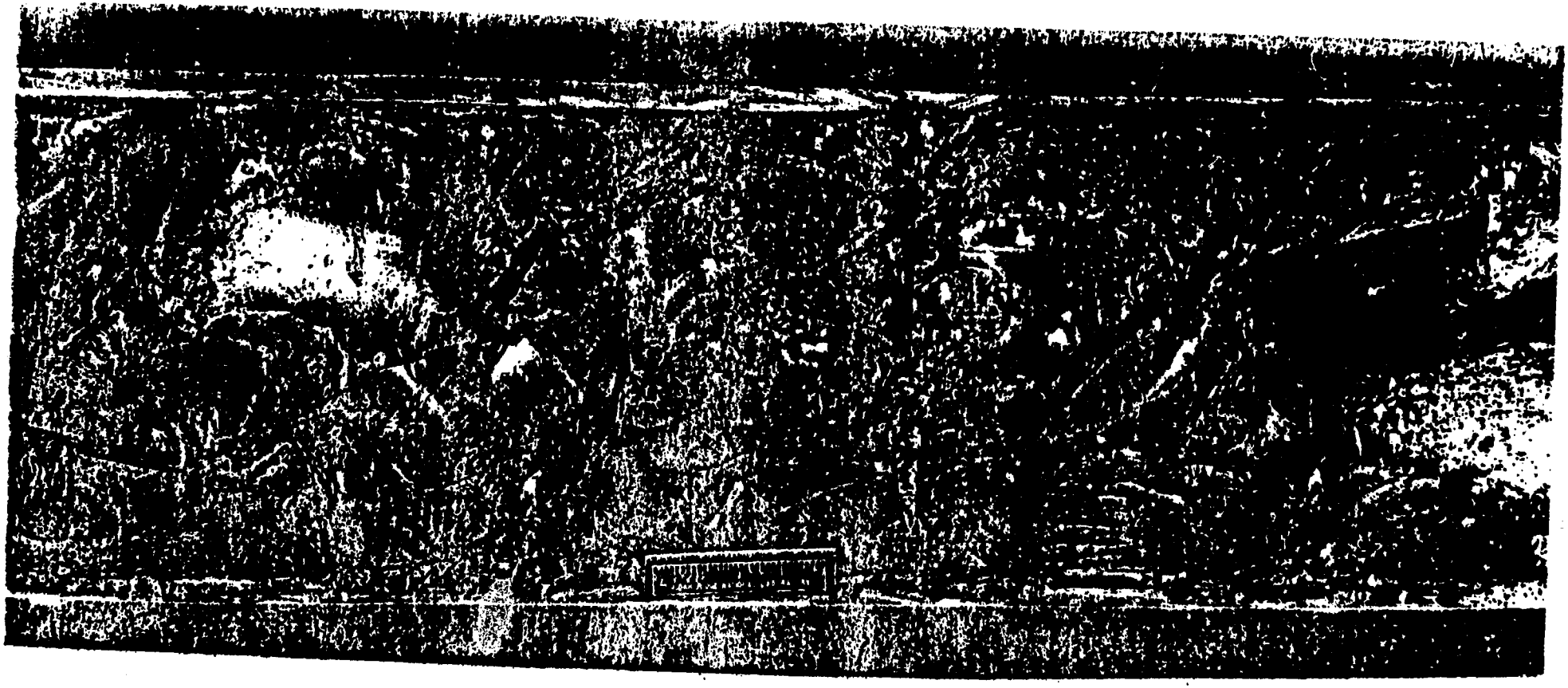
GLASS

SYNROC

SUPERCALCINE

CERMET

MULTIBARRIER



Radioactive Glass Prepared From TBP-25 Aluminum Waste