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Glass Science Tutorial Lecture # 5: Historical Review of USDOE Tank Waste Management

Prepared for the U.S. Department of Energy
Office of Environmental Restoration and
Waste Management

Westinghouse
Hanford Company Richland, Washington

Hanford Operations and Engineering Contractor for the
U.S. Department of Energy under Contract DE-AC06-87RL10930

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E. W. McDaniel, Lecturer

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HISTORICAL REVIEW OF USDOE TANK WASTE MANAGEMENT

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A SHORT COURSE

PRESENTED FOR WESTINGHOUSE HANFORD COMPANY

FEBRUARY 15-16, 1995

HISTORICAL REVIEW OF USDOE TANK WASTE MANAGEMENT

EARL W. MCDANIEL

Chemical Technology Division
Oak Ridge National Laboratory

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1. OBJECTIVE

The objective of this two-day short course is to present an unbiased historical overview of the DOE tank waste activities. Material covered is readily available in the open literature. There is no intent to be critical of past practices. As I proceed through the course I will inject world events that impacted the United States nuclear program or, in some cases, how world events were impacted by the United States Nuclear program.

As the audience is well aware, both solid and liquid waste results from most nuclear operations. In my discussion I will confine myself to liquid, mostly tank waste, and sludge. We will start out at the dawn of the nuclear age and end up at the Hanford Site in 1995.

CHRONOLOGY

1895	Wilhelm Roentgen discovers X-ray radiation.
1932	James Chadwick discovers the neutron.
1938	Otto Hahn and others achieve atomic fission.
October 1939	Albert Einstein and others persuade FDR to support research on an atomic bomb.
December 1941	FDR approves the Manhattan Project.
December 1942	Enrico Fermi and others achieve controlled and sustained chain reaction.
July 1945	First atomic bomb detonated at Alamogordo, New Mexico.
August 1945	Hiroshima and Nagasaki are destroyed by atomic bombs.
June 1946	U.S.A. proposes Baruch Plan for international control of nuclear weapons; U.S.S.R. rejects it.
June 1946	First Atomic Energy Act enacted.
August 1949	U.S.S.R. detonates its first fission bomb.
January 1950	President Truman approves development of the hydrogen bomb.
May 1950	NSC-68 outlines plan for massive U.S. rearmament and military spending.
January 1951	AEC begins weapons testing at Nevada test site.
December 1951	First experimental production of electricity from a nuclear reactor.
November 1952	U.S.A. detonates its first hydrogen bomb, followed by U.S.S.R. in 1953.
December 1953	President Eisenhower's Atoms for Peace address.
January 1954	John Foster Dulles announces a new strategic policy, massive retaliation.
June 1954	AEC affirms denial of security clearance to J. Robert Oppenheimer.
August 1954	Second Atomic Energy Act enacted.

September 1957	Congress enacts the Price-Anderson Act, indemnifying the nuclear power industry in the event of nuclear accidents.
October 1957	U.S.S.R. launches Sputnik. The missile gap is born.
December 1957	Shippingport, the world's first exclusively non-military power reactor, comes on stream.
? 1957	U.S.S.R. nuclear disaster near Sverdlovsk.
Spring 1958	Worldwide protests begin against atmospheric nuclear testing.
January 1961	President Kennedy advances a new strategy, flexible response.
June 1962	Robert McNamara announces No Cities doctrine.
July 1963	U.S.A., U.S.S.R., U.K. sign Partial Test Ban Treaty, ending atmospheric testing.
February 1964	Mutually assured destruction becomes the nuclear strategy of U.S.A.
October 1966	Nuclear accident at Fermi-1, a commercial breeder reactor near Detroit.
September 1967	U.S.A. announces it will deploy a "thin" antiballistic missile system.
January 1972	AEC generic rule-making hearings begin on ECCS.
May 1972	SALT I is signed in Moscow. ABM deployment is sharply limited.
August 1974	Rasumussen report released, downplaying likelihood of reactor accidents.
October 1974	Congress abolishes AEC, substituting NRC and ERDA (which is later replaced by Department of Energy).
November 1974	Karen Silkwood killed in accident.
November 1974	U.S.A., U.S.S.R. agree to Vladivostok accords.
January 1975	Browns Ferry reactor accident.
March 1979	Three Mile Island; concurrent, but coincidental release of film, *The China Syndrome.*
June 1979	SALT II signed in Moscow.
December 1979	SALT II withdrawn from Senate by President Carter.

2. INTRODUCTION

As the course progresses I will make a distinction between high-level and other types of liquid waste, but for the present I will use the term "tank waste" to indicate liquid waste that has resulted from the processing of fuel elements.

Chronologically, the first waste of this nature was generated in the Manhattan Project of World War II. The Atomic Energy Commission, now the Department of Energy (DOE), which was the outgrowth of the Manhattan Project, pursed two simultaneous courses after the war. The first was, (1) The United States national defense plans called for the production of plutonium, while on the other hand, a program for peaceful use of atomic energy promoted an increasingly sophisticated nuclear technology devoted to the production of electricity. A product common to both activities is the generation of highly radioactive wastes. It should be noted at this point that the waste generated takes a variety of forms and there is no conveniently simple definition. However, as I progress through the course, waste classifications will be discussed.

The management (storage, treatment, and disposal) of radioactive liquids has historically been one of the major problems facing the nuclear industry. Besides being a new problem, in the early days on the industry, it has certain novel features. For example, it is impossible by any known chemical or physical means to destroy the property of radioactivity. In addition, in today's terms, many impurities in the fuel and processing equipment has resulted in a waste also contaminated with hazardous components.

The tank wastes, for the most part, come from the processing of spent fuel. Uranium fissions into some 30 radionuclides, or fission products. From a chemical point of view, fission products represent a very wide assortment of elements. The exact proportions depend on the type of fuel used, on the rate of burn-up in the reactor, and the time elapsed from time of removal to processing.

Typical fission products for one ton of uranium is given in Table 1. Let me walk you through a generic processing scheme. First, the irradiated fuel elements are dissolved in nitric acid and a

solvent extraction procedure carried out. The uranium and plutonium are extracted in the organic phase and the separated fission products remain in the aqueous faction. The level of activity of such liquids averages 10 to 15 curies a liter.

Table 2 shows a typical mixture of wastes resulting from processing 1 ton of irradiated fuel. In general, in the United States, the wastes are neutralized and then stored in mild steal tanks.

Table 1. Fission products resulting from the burn-up of 1 ton of uranium.

Chemical Group	Chemical Element	Weight (kilograms)
Rare gases	Krypton and Xenon	128
Heavy alkalis	Rubidium	15
	Caesium	118
Alkaline earths	Strontium	42
	Barium	43
	Rare earths and Yttrium	317
Group 4 elements	Zirconium	125
Group 5 elements	Niobium	5
Group 6 elements	Molybdenum	92
	Tellurium	16
Group 7 elements	Technetium	29
	Iodine	7
Rare metals	Ruthenium, Rhodium, and Palladium	61

Table 2. Typical mixture of wastes arising from processing of 1 ton of irradiated fuel.

Chemical	Weight (tons/10,000 metre^3)
Nitric acid	1,300
Ferric nitrate	7
Chromium, aluminum, nickel, and uranium nitrates	5
Mixed fission product nitrates	1-8

A Selection of World Events That Impacted Nuclear Weapons Development.

1. Potsdam Conference, July, 1945
2. Atomic bombing of Japan, August, 1945
3. David Greenglass, a machinist at LASL, works on implosion lens of Pu bomb.
4. Russians block surface access to Berlin, 1948—49
5. Russia explodes first nuclear device, Aug., 1949
6. Korean conflict starts June, 1950
7. Savannah River Site starts producing nuclear materials, 1953
8. Thermonuclear weapons development accelerates
9. Gaseous diffusion facilities expanded
10. Additional reactors built at Hanford
11. Rocky Flats Plant built, Dow becomes operator 1953
12. McCarthy with hunt starts, 1950
13. Julius and Ethyl Rosenberg, atomic spies electrocuted June, 1953
14. Birth of Nuclear Navy, 1950
15. Light water reactors commercialized, 1956
16. Atoms for peace program, 1953
17. John Foster Dulles offers French nuclear weapons to use in Indo-China, 1954
18. Camp Century in Greenland built, 1960
19. U-2 incident, 1960
20. John F. Kennedy assassinated, 1963
21. China explodes nuclear device, 1964
22. USSR breakup, 1991

MEETING AT POTSDAM

JULY 17 TO AUGUST 2, 1945

For two weeks in the summer of 1945, Harry S. Truman, Winston Churchill, and Josef Stalin gathered at Postdam, a suburb of Berlin, to reconstruct the world out of the ruins of the Second World War. The post war arms race started at this meeting.

The United States had been very careful not to inform the Russians about the development of Atomic Energy for military purpose. Many American military officials, especially General Groves, head of the Manhattan Project, considered Russia the real enemy when it came to atomic secrets. When Mr. Truman became president upon the death of Mr. Roosevelt he was briefed on the Manhattan Project for the first time. In Mr. Truman's mind there was never any question about using the bomb when one was ready.

During the Potsdam conference Mr. Truman was informed of the successful Trinity test. Question was what and how much to tell Josef Stalin. Mr. Truman decided to tell him after one of their meetings that the U.S. has developed an entirely novel form of a bomb, something quite out of the ordinary (without mentioning the word atomic), which we think will have a decisive effect upon the Japanese's will to continue the war. On July 22, 1945, President Truman casually told Stalin that America had perfected a very powerful explosive. Stalin was just as casual, saying that he hoped America would make good use of the weapon against Japan. Stalin's impassive reaction lead most present to assume that Stalin had not grasped the importance of Truman's announcement. Clearly he had, if only because Soviet agents had kept him informed of America's bombs. Marshal Georgi Zhuhov wrote in his memoirs that "on returning to his quarters, Stalin told Molotov about his conversation with Truman. The Molotov reacted immediately saying, "let them, we'll have to talk it over with Kurchatov and get him to speed things up." Thus, started the arms race.

BERLIN BLOCKADE

Politics and geography made Berlin the gravest and most persistent source of tension in the nuclear age. Post war Berlin, like Germany itself, was split into four zones of occupation. France, Briton, the U.S., and the Soviet Union each had a sector which was to last for nearly 50 years. Berlin was located 110 miles into Soviet-controlled East Germany.

Berlin tempted the Soviets because their rivals were at a disadvantage in protecting their access to the city. A failure to do so would be a great blow to the Western credibility, especially that of the Americans.

Let me now shift my story to Los Alamos Scientific Laboratory. The Atomic Energy Commission had recently been established and assumed control of existing nuclear weapons; four to be exact, which were stored at Los Alamos. A routine inspection showed that the weapons were useable due to some over looked storage problem. All Pu would have to be removed and reworked. It was expected to take about a year, but within 72 hours of this discovery, the Soviets sealed off Berlin by a total blockade. The date was June 24, 1948. The blockade would last until May 12, 1949. It ended a few weeks before the Americans again had atomic weapon capability.

During the blockade, the U.S. Air force sent two squadrons of B-29s of the same type that was used to drop the two atomic bombs on Japan, but the Soviets were not moved. Later in 1949 the Soviets would test their own plutonium atomic bomb.

On June 25, 1950, Soviet backed North Korea invaded South Korea, thus adding fuel to the Nuclear arms fire. The Korean Conflict, as it was then called, was to last until July 27, 1953.

During this period the nuclear weapons complex experienced massive growth.

1. The Savannah River Plant began producing nuclear materials in 1953.
2. In 1953 Rocky Flats Plant became operational.
3. New gaseous diffusion plants were built.
4. H—bomb development accelerated.

MAJOR WORLD WAR II SITES

The major Manhattan Project sites of World War II were:

1. The Hanford Engineering Works, Richland, WA
2. Clinton Engineering Works, Oak Ridge, TN
3. Los Alamos Scientific Laboratory, Los Alamos, NM
4. Columbia University, Upton, LI, NY
5. University of Chicago Metallurgical Lab., Chicago, IL

In addition to these major sites, there were a large number of minor sites providing support services. I will discuss the first three. Hanford and the Clinton Engineering Works produced tank waste in some proportion. The other sites did not generate fuel processing wastes. Sites that were built in the 1950/60s that will be discussed are:

1. Savannah River Site, Akin, SC
2. National Reactor Test Center, Idaho Falls, ID
3. West Valley Nuclear Services, West Valley, NY
4. Rocky Flats Plant, Golden, CO

CLINTON ENGINEERING WORKS

OAK RIDGE, TN

Clinton Engineering Works was the original name of the present Oak Ridge Site. I am not sure what the politically correct name is. Facility or site names seem to change with the Secretary of Energy, but I hope you know where I am talking about. The site of the Clinton Engineering Works was selected by General Leslie Groves due to its remote location and the availability of cheep electrify and an abundant water supply. The site is located on the Clinch River some 25 miles west of Knoxville. The name is taken from the name of Clinton, the county seat of Anderson County in which the city of Oak Ridge is located and one of the plants. The name Oak Ridge was derived from one of the ridges that naturally contained a large number of oak trees. There was no town of Oak Ridge prior to the Manhattan Project. There were several small farming communities such as Wheat and Robertsville. I live on Robertsville Road. The local residents were moved out to make room for the project. For some families this was the third time the U.S. Government had relocated them. Some from the Great Smoky Mountain National Park, some the Tennessee Valley Authority and now some mysterious project that John Hendrix had a vision about some 20 years earlier. The cheep electricity was from Norris Dam, the first TVA dam, about 27 miles up stream on the Clinch River. Clinton Engineering Works was divided into 3 distinct activities:

1. Clinton Laboratories, now Oak Ridge Laboratory, at one time Hollifield National Laboratory coded, the X-10 site.
2. Y-12 Electromagnetic Plant, this plant was coded Y-12.
3. The Gaseous Diffusion Plant, coded K-25. Some say site designators were taken from map coordinates. However, your guess is as good as mine as to where they really came from.

These plants are each located in a separate valley .

CLINTON LABORATORIES (X-10)

Clinton Laboratories was the site of the Graphite Pile, as it was called then. The reactor was built as a pilot plant for the Hanford reactors. It was fueled with natural uranium, graphite moderated and air cooled. Its purpose was to produce Pu in gram quantities. The reactor, which operated until 1963, is now a historic site. Located adjacent to the reactor was the world's first reprocessing plant, which was really a pilot plant to develop and evaluate methods for separating uranium and plutonium. The acid waste was neutralized and stored in gunite tanks, which I will discuss in some detail when I discuss hydrofracture development and operations at Oak Ridge National Laboratory. Another mission of the laboratory was to evaluate the possibility of separating uranium 235 via thermal diffusion. After the war peaceful uses of the atom were explored and ORNL became the world's major producer and supplier of radio and stable isotopes. The Y-1 calutrons were modified to separate a wide variety of mass numbers. In the early 50s we saw the birth of the Nuclear Navy. Admiral Richover and a group of naval officers attended the Oak Ridge School of Reactor Technology (ORSORT). During this period we saw the birth of the light water reactor. According to Milt Shaw, all theoretical reactor development was complete and future funding would be devoted to engineering. Al Weinberg, ORNL Director and reactor expert, saw the light water reactor as an interim source of energy. Dr. Weinberg was looking at breeders, especially the thorium fuel cycle. This cycle bred Th-232 into U-233, which was fissionable. To quote Dr. Weinberg "electricity will be too cheep to meter." Thus started a long term feud between Weinberg and Shaw, which terminated in 1971 when Dr. Weinberg resigned as ORNL director.

This period of civilian reactor development, perhaps described as the Golden Era, was a very exciting time to be associated with the nuclear industry. Development engineers and scientists were so excited and enthusiastic about their work they were practically living at the lab. Dr. Shipley, one of the pioneers of fusion energy actually had cots brought in so his staff would have a place nearby to nap. A great deal of this activity was in support of President Eisenhower's atoms for peace program.

Project plowshare came into being. This project explored the use of nuclear explosives to enlarge harbors for larger nuclear powered ships, build a new canal to replace the Panama Canal. In 1960, Camp Century came into being. This was an army base built under the ice in Greenland and all power was powered by a small reactor. The Aircraft Nuclear Pulpulsion (ANP) project was born but never really got off the ground. Neither did the atomic powered rocket. These were some of the activities that were happening that required more and more fissionable materials, both plutonium and highly enriched uranium. These activities coupled with advanced weapons systems development were used to justify the SRP, additions to the Hanford Site and the building of new gaseous diffusion plants in Kentucky and Ohio. In producing all this nuclear material, many gallons of tank waste were produced. Contrary to popular opinion, there was always serious thought given to managing these highly active tank wastes and sludges. In 1959, Oak Ridge National Laboratory embarked upon developing hydraulic fracturing as a method of terminal disposal of liquid wastes and later sludges. I will discuss this development in detail later.

I WILL NOW SHIFT BACK TO THE TOPIC OF DISCUSSION - TANK WASTE

As I stated in my introductory remarks that at the end of WWII the United States followed two nuclear paths: (1) continued development of atomic energy for military purposes and (2) development of peaceful uses of the atom, i.e, power reactor development. Westinghouse and General Electric became giants in the industry, marketing light water reactors all over the western world. We saw the French, British, Germans, USSR, and later Japan, Korea, Taiwan and mainland China opt for nuclear power. Little thought appeared to be given about the waste generated in providing fuels. Weapons production waste was cloaked by national security. If you wanted to keep something from the public, your boss, or a fellow worker, you just classified it. Not as much work on the civilian fuel cycle could be cloaked. Thus, the public became active on this side on nuclear energy not knowing that the AEC was the world's major producer of nuclear waste.

The disposition of radioactive waste from the nuclear electric power industry continues to be a subject of much interest to the public, and in particular, to those searching for any possible vehicle to stop or delay the nuclear power program. In recent years, the emphasis has shifted from the nuclear utilities to the cleanup of such sites as Hanford, Oak Ridge, Savannah River, and Rocky Flats. But as you are well aware, the nuclear utilities industry in this country is on its death bed. The Japanese, French, Koreans, Taiwan, and Argentina are still aggressively pursuing a nuclear power program. The attitude developed during this era that we do not know how to permanently manage the waste and until we know how, no more should be produced. I will not get into this topic, but will mention it.

Y-12 PLANT ELECTROMAGNETIC PLANT

The Y-12 Plant is still called the Y-12 plant. The plant, built largely on the prestige of Dr. E. O. Lawrence, inventor of the cyclotron, was to separate uranium isotopes electromagnetically. Dr. Lawrence modified his cyclotron into an isotope separator and called it a calutron. General Groves did not have much confidence in the process, but felt it would be good to leak information to the Russians, whom Groves considered the real enemy, and keep them away from gaseous diffusion, which General Groves was sure would be able to enrich large amounts of uranium once in production. It should be noted that the Hiroshima bomb was made from uranium separated at Y-12. The Plant was composed of 4 beta process buildings and 5 alpha process buildings. The beta buildings contained calutrons that had a 24—inch radius and were numbered 9204—1, 2, 3, and 4. The alpha buildings had 48—inch radius calutrons and were numbered 9201—1, 2, 3, 4, 5. Wonder what the 92 meant? Silver from Fort Knox was used to wind the electromagnetic coils since copper was a strategic material. Many tons of silver was used. The last was removed and returned to the U.S. Treasury in 1969. General Groves once described calutrons as the most inefficient process ever put into mass production. Efficiency was between 5 and 15%. After the war, most of the process buildings were striped of the calutrons and Y-12 became the workhorse of the nuclear arsenal playing a major role in H—bomb development. The remaining calutrons were converted for use in separating stable isotopes. After 50 years one building still exists. Y-12 is an interesting place. It has had a major role in all U.S. weapons systems. Y-12 in cooperation with K-25 was very active in the nuclear navy weapons systems. It is the site of the lithium separations for thermonuclear weapons development in the early 1950s. While X-10 continued to play a major role in developing the civilian nuclear fuel cycle and became a major site of reactor development. During the lithium operations, the AEC cornered the world market on elemental mercury and controlled the lithium market. Lithium was amalgamate with mercury and isotope separation was done in an electric cell. The lithium 7, which was unwanted was converted back to carbonate and returned to the Lithium

Corporation of America, Bessemer City, NC for sale on the open market. As the world major lithium deposits were owned by the Lithium Corporation of America if you wanted lithium you had to buy it from them. This is how the AEC found out that the French were developing H—bombs. The French were buying lithium from the Lithium Corporation and were trying to separate the isotopes. The French were, in fact, being sold depleted lithium. When the French complained to the Lithium Corporation they inturn passed this information on to the USAEC. Also of historical note, Dr. H. H. Willard, for many years professor of chemistry at the University of Michigan and consultant to the AEC, determined the atomic weight of the newly discovered element, lithium for his PhD research at Harvard around 1903. His atomic weight was not accepted by the scientific community until the early 1950s, when the Lithium Program at Y-12 confirmed his determination to 6 decimal places. He never tired of telling this story.

GASEOUS DIFFUSION PLANT (K-25)

Gaseous diffusion was developed by Airy and his team at Columbia University. This was the birth of Brookhaven National Laboratory, by the way. Gaseous diffusion diffuses UF6 through a series of nickel barriers until the desired enrichment is achieved. The original diffusion building was designated K-25. This building was at one time the world's largest building under a single roof. The building was "U" shaped and was one mile around the building. Much of the gaseous diffusion information still is classified. Several additional gaseous diffusion buildings incorporating advanced technology was built in the late 40s and early 50s, as were the plants at Paducah, Kentucky and Portsmouth, Ohio. Large amounts of electrical energy was required for the gaseous diffusion process that a large number of scientists estimated that based on the amount of energy required to enrich uranium to 2-3% to use as power reactor fuel, these reactors were not all that energy efficient. The K-25 Site is now an Environmental Management Site and home of Oak Ridge's TSCA incinerator. Much good work came out of K-25. The site supported the development of teflon for use with fluorides. As nickel metal was inert to fluorene and fluorides, much nickel metallurgical development was required, most of which was done either at K-25 or in support of K-25. Of particular interest is the powder metallurgical techniques developed in support of the barriers. Also, many parts were nickel plated.

HANFORD ENGINEERING WORKS

As you are all Hanford Site employees, I assume you have some detailed knowledge of the birth and development of the activities here so I will not go into great detail. I am sure you have a greater knowledge of the site than me. I have been associated with Hanford since 1978 and been coming here on a regular basis since October, 1981. Later I plan to discuss in some detail the history of the late Hanford Grout Program. For now I will say a few words about the waste. Radioactive wastes have accumulated at the Hanford site since 1944, when the first reactor fuel was processed for plutonium recovery. High-level liquid waste, generated by the Purex and BiPO4 processes have been stored as neutralized slurries in over 150 underground storage tanks. In 1957, a program was started to insure safe containment of these liquid wastes. This early effort developed into the Hanford Waste Management Program, which started in about 1968 and was to be completed in 1980. The chief goal of the program was to assure isolation of radioisotope from life forms. During this period a big effort was devoted to removing cesium and strontium from the tank wastes. I think I will stop here and pick it up later when I talk about the history of the grout program.

ROCKY FLATS PLANT

Rocky Flats Plant is located in Jefferson County, CO on State Highway 93 about one-half way between Golden and Boulder. It was built in the 1950s and operated by Dow Chemical Company, later by Rockwell International, and now by EG&G. In the late 1970s to early 1980s, a reprocessing plant was built at RFP. Due to some major design flaws, the plant never became operational. I have a short video that describes the plant.

LOS ALAMOS SCIENTIFIC LABORATORY

Los Alamos Scientific Laboratory, now Los Alamos National Laboratory, was born out of necessity in 1943. J. Robert Oppenheimer, the labs first director, picked this remote site in NW New Mexico about 35 miles NW of Santa Fe. People checked into a post office address in Santa Fe. Oppenheimer was familiar with the site having spent summers in the area at a boy's ranch. It is a very remote location and is composed of a series of canyons and mesas. Los Alamos National Lab is composed of technical areas, which compounds some several miles from the main laboratory. Los Alamos Scientific Laboratory is significant in that this is where the uranium from Oak Ridge and plutonium from Hanford were turned into weapons. In addition, to bomb physics, many metallurgical problems were to be overcome in producing acceptable weapons grade metal. In this activity much uranium and plutonium were redissolved and reprocessed with much liquid waste resulting. The staff at Los Alamos were so sure that the uranium bomb would work that it was not tested until it was dropped over Hiroshima. The major problem with the plutonium bomb was developing correct implosion techniques using conventional explosives. David Greenglass, brother of Ethyl Rosenberg and a machinist, worked on the implosion lens. This was the information passed on to the Russians and the crime for which Julius and Ethyl Rosenberg were electrocuted. David Greenglass received a 30—year sentence for his part. The engineering arm of Los Alamos Scientific Laboratory was located in Albuquerque. It became a separate laboratory in 1948 being named Sandia Laboratories, operated by AT&T until 1993, when Martin Marettia Corporation assumed management responsibility. Los Alamos Scientific, now Los Alamos National Laboratory, has been operated by the University for the past 51 years. Atomic weapons continued to be designed and built at LANL for several years after the war. Weapons were stored either at Los Alamos or at the Sandia Base in Albuquerque. In the early 50s Rocky Flats was built as the manufacturing facility for plutonium weapons parts. Los Alamos continued to design and test nuclear devices until the early 1990s

NATIONAL REACTOR TESTING STATION

IDAHO FALLS, ID

The name of the National Reactor Test Station was changed to Idaho National Engineering Laboratory when the Atomic Energy Commission created the NationaL Laboratory system. Most of the highly active liquid waste at the INEL is generated at the Chemical Processing Plant (CPP) during the reprocessing spent nuclear fuel. Unlike the Savannah River and Hanford sites, the CPP reprocesses fuel made from highly enriched uranium. The highly active waste is mainly in the first cycle raffinate from the solvent extraction of the dissolved fuel solutions. The waste is self-heating in both solid and liquid forms. Although the second and third cycles are not self-heating, at the CPP these wastes are consider to be high-level using the definition that any waste that has been in direct contact with the fuel element is high-level. The highly active liquid wastes are stored in a tank farm consisting of eleven 300,000 gal stainless steel tanks located in underground concrete vaults awaiting calcination. The waste normally was stored about 4 years prior to calcination. The policy at the CPP has been to calcine the liquid waste and to store the granular solids in engineered bins where the waste is fully retrievable. The Waste Calcining Facility (WCF) started calcining highly active liquid waste in a fluidized-bed calciner in December, 1963.

HISTORICAL DEFINITIONS ON HIGH-LEVEL WASTE

In the earliest descriptions of high-level waste (HLW), the term "high-level" often was associated with two attributes of the waste: (1) high levels of external radiation that would necessitate extensive shielding to protect workers during waste handling and (2) high levels of heat from radioactive decay that would necessitate engineering systems for heat removal, i.e., to prevent self-boiling or self-dispersal of the waste. High levels of external radiation and decay heat resulted principally from high concentrations of shorter-lived fission products. The earlier descriptions of HLW thus were related only to the need to control short-term risks from waste handling and storage, but the descriptions did not consider attributes of the waste related to control of long-term risks from final disposal.

In addition to the descriptions of HLW in terms of high levels of external radiation or decay heat, the concept was developed that HLW is waste of certain origin, i.e., from chemical reprocessing of spent nuclear fuel, because this was the only known source of waste with these properties. Thus, HLW came to be regarded as waste from fuel reprocessing in which most of the shorter-lived fission products have not decayed and significant radionuclide separations or waste dilutions have not occurred.

The AEC also provided some definitions of commercial high-level waste, but I will skip over his and go to the description given in the Waste Valley Demonstration Project of 1989 (Public Law 96-368). The act includes the following definition:

> The term, "high-level radioactive waste," means high level radioactive waste which was produced by reprocessing at the (West Valley) Center of spent fuel. Such a term included both liquid wastes, which are produced directly in reprocessing, dry solid materials derived from such liquid waste, and such other material as the (Nuclear Regulatory) Commission designates as high level radioactive waste for purposes of protecting the public health and safety

The NRC has not yet designated any "other material" as HLW under the West Valley Act. Rather the NRC has interpreted this term in a manner consistent with the definition in 10 CFR part 50. HLW is the liquid wastes in storage at West Valley and the dry solid materials derived from solidification of the liquid wastes. In NRC terms, "high-level radioactive waste means, (1) irradiated reactor fuel, (2) liquid wastes resulting from the operation of the first fuel cycle solvent extraction system and the concentrated wastes from subsequent extraction cycles in a facility for reprocessing irradiated reactor fuel, and (3) solids into which such liquid wastes have been converted." Later we will see a modification of this definition in the Nuclear Waste Policy Act of 1982.

U.S. ENVIRONMENTAL PROTECTION AGENCY

The EPA's 40 CFR Part 191 contains generally applicable environmental standards for the management and disposal of spent fuel, HLW, and as yet unmentioned TRU waste. These standards apply not only to commercial wastes, the disposal of which would be licensed by the NRC according to the technical criteria in 10 CFR Part 60, but also to the DOE's defense wastes. EPA defines spent fuel as fuel that has been withdrawn from a reactor following irradiation, but has not yet been reprocessed. HLW is defined as in the Nuclear Waste Policy Act, which I will discuss in a few minutes.

U.S. DEPARTMENT OF ENERGY (USDOE)

The definition of HLW currently used by the DOE is contained in Order 5820.2 and is similar to those of the NRC and EPA. HLW is defined as follows:

> "The highly radioactive waste material that results from the reprocessing of spent nuclear fuel, including liquid waste produced directly in reprocessing and any solid waste derived from the liquid that contains a combination of TRU waste and fission products in concentrations as to require permanent isolation."

In the NWPA, HLW is defined as:

(a) "the highly radioactive material resulting from the reprocessing of spent nuclear fuel, including liquid wastes produced directly in reprocessing and any solid material derived from such liquid waste that contains fission products in sufficient concentrations; and (b) other highly radioactive material that the Nuclear Regulatory Commission, consistent with existing law, determines by rule requires permanent isolation."

It should be noted at this point that the definition of HLW in the NWPA does not apply to the DOE's defense waste unless commercial and defense waste are mixed.

DEFINITIONS OF TRANSURANIC WASTE (TRU)

U.S. Environmental Protection Agency. In the EPA's 40 CFR Part 191, TRU waste is defined as:

> "... waste containing more than 100 nanocuries of alpha-emitting transuranic isotopes, with half-lives greater than 20 years, per gram of waste, except for: (1) high-level radioactive waste; (2) wastes the Department of Energy has determined with concurrence of the administrator, do not need the degree of isolation required by this part; or (3) wastes that the NRC has approved for disposal on a case-by-case basis in accordance with 10 CFR Part 61.

U.S. Department of Energy. In DOE Order 5820.2, TRU waste is defined as:

> "Without regard to source or form, radioactive waste that, at the end of institutional control periods is contaminated with alpha-emitting transuranium radionuclides with half-lives greater than 20 years and concentrations greater than 100 nCi/g.

The definition of TRU waste is thus the same as that of the EPA and the definition also is explicit in excluding HLW and spent fuel. I did not go into this part of the definition. U.S. Nuclear Regulatory Commission. The NRC's 10 CFR Part 61 does not explicitly define TRU waste, but the standards set a concentration limit of 100 nCi/g for the general acceptability of near-surface land disposal for alpha bearing TRU radionuclides with half-lives greater than 5 years.

LOW-LEVEL WASTE

This is the classification we are all perhaps most interested. I have took a long time getting here. Current definitions of LLW differ from those for HLW and TRU in the sense that LLW is defined by exclusion. DOE Order 5820.2 defines LLW as "radioactive waste not classified as high-level, TRU, or spent fuel.

I am sure you all are aware that I have just highlighted the cited regulations and there is much more to them than what is stated here. Most of what I have discussed is of about 1986-87 time frame. Also, I have not even mentioned mixed waste.

As for the NRC, low-level wastes are classified in 10 CFR Part 61 only in relation to risks associated with waste disposal, but risks associated with waste disposal operations have no bearing on the concentration limits in the standards.

DOE Order 5820.2 contains explicit provision that LLW shall be disposed of by shallow-land burial or greater confinement disposal (GCD). It is my understanding that DOE Order 5820.2 has been revised and has been circulated at least in DRAFT, however, I have not seen the DRAFT.

Thus, as with HLW and TRU waste, these definitions and descriptions of LLW do not associate these wastes with a particular disposal technology.

MIXED WASTE

I will close out this section on mixed waste. This topic is very complicated and in many cases ill defined. EM-50 has pumped many millions of dollars into the Mixed Waste Integrated Program for the past few years to work on this topic. For the purposes of our discussion we will say that all the prior definitions are applicable and with the presence of certain hazardous chemical as listed by the EPA the waste becomes "mixed" and a new set of rules apply.

WEST VALLEY NUCLEAR FUEL SERVICES (NFS)

WEST VALLEY, NY

The West Valley site was the location of the only operating commercial nuclear fuel reprocessing plant in the United States. NFS operated this facility from 1966 until 1972, processing 640 metric tons of commercial and defense fuels using the PUREX process. It should be noted that the Atomic Energy Commission promised NFS a base load. In fact, some N-reactor fuel was processed at West Valley. Between 5 and 6 million gallons of fuel reprocessing waste resulted from this operation. Most of this waste was stored in an underground storage tank. The bulk of the tank's contents resulted from the addition of excess sodium hydroxide to neutralize the nitric acid stream resulting from the first solvent extraction cycle.

The West Valley Demonstration Project Act

On October 1, 1980, Public Law 96-368 directed the U.S. Department of Energy (USDOE) to carry out a high-level radioactive (HLW) demonstration project at the former Nuclear Fuel Services site. West Valley Nuclear Services (WVNS), a subsidiary of Westinghouse Electric Corporation, was chosen to be the prime contractor, and assumed control of the site in February, 1982. Early in the project, two decisions were made that determined the major thrust of the HLW solidification effort:

1. The HLW alkaline supernate would be separated from the sludge, and the radioactive components in the supernate would be chemically separated and combined with the sludge into a terminal waste form. The treated supernate would be processed into a suitable low-level waste form.
2. The terminal HLW form would be borosilicate glass. This second decision was consistent with the Hinch Committee recommendation in 1981 that all HLW be vitrified. The only unanswered question was what was a practical definition of HLW that everyone could and would agree to.

BACKGROUND FOR DOE DECISIONS FOR THE DISPOSITION OF THE HLW

After the West Valley Demonstration Project (WVDP) started, the Department of Energy had to decide how they wanted the waste to be handled. There were several alternatives considered as approaches to solidify the HLW into a form suitable for disposal in a federal repository.

1. On-site Processing to a Terminal Waste Form
    A. Separated salt\sludge process-remaining high-level radioactive components after decontamination of supernatant mixed with HLW sludge
    B. Nonseperated salt\sludge—no separation or concentration
2. On-site processing to an Interim Waste Form/Liquid HLW converted to solid interim waste form for future processing to terminal form
3. In-tank solidification/HLW mixed with cement and poured into existing tanks
4. No action

Alternative 1 was the only one that complied with the West Valley Demonstration Act requirement to solidify liquid HLW into a form suitable for transportation and disposal at West Valley Alternative 1a was selected on the basis that it appeared to be more environmentally acceptable. As a result of this decision by the DOE, The Sludge Treatment System (STS) was created to decontaminate the supernate for on site treatment. In all, eight systems were constructed for the treatment of the HLW and LLW.

SYSTEMS BUILT TO TREAT HLW AND LLW AT WVDP

1. CEMENTATION SOLIDIFICATION SYSTEM (CSS)
2. LIQUID WASTE TREATMENT SYSTEM (LWTS)
3. SUPERNATANT TREATMENT SYSTEM(STS)
4. WASTE IMMOBILIZATION SYSTEM (WMS)
5. VITRIFICATION SYSTEM (VS)
6. HIGH LEVEL WASTE INTERIM STORAGE
7. SIZE REDUCTION FACILITY
8. TRANSURANIC ASSAY AND INTERIM STORAGE

The objective of the supernatant decontamination was to separate the majority of the radioactive species from the nonradioactive salt in order to minimize total volume of HLW waste glass. The major species being cesium. The supernatant is further processed in the LWTS and finally sent to the CSS for encapsulation in cement and disposal. The remaining HLW is sent to the VS where it will be solidified in borosilicate glass for eventual disposal in a federal repository.

During 1983 and 1984, many cesium removal processes were evaluated. It was discovered that the cesium could be removed using a zeolite which could be melted into glass without adverse effects. Direct melter feed would permit simplification of the STS.

Precipitin methods were considered at WV as they had been used at other DOE sites.

PRECIPITIN METHODS CONSIDERED

1. Ferrocyanide
2. PTA
3. NaTBP

The highest decon factors were obtained using IE-95/96. It is worth noting that IE-95 was successfully used at Hanford in the 1950s to remove Cs from PUREX supernatant. In addition to the West Valley facility, reprocessing plants were built by the following:

1. General Electric's Midwest Fuels Reprocessing Plant at Morris, IL.
2. Allied General Nuclear Services, at Barnwell, SC
3. A plant to be operated by Exxon Nuclear Corp. was scheduled to be built in Tennessee.

None of the above plants ever operated.

Just to name a few major activities. I will now discuss in some detail:

1. The Oak Ridge National Laboratory's Hydrofracture Program
2. The Savannah River Saltstone Effort.
3. A brief history of the Hanford Grout Program

SAVANNAH RIVER PLANT

The Savannah River Plant occupies an area of approximately 300 square miles along the Savannah River near Aiken, SC, approximately 22 miles down stream from Augusta, GA. The plant includes a nuclear reactor fuel fabrication plant, at one time 5 reactors operated, two reprocessing plants and a facility for producing heavy water. The plant started producing nuclear materials for the AEC in 1953.

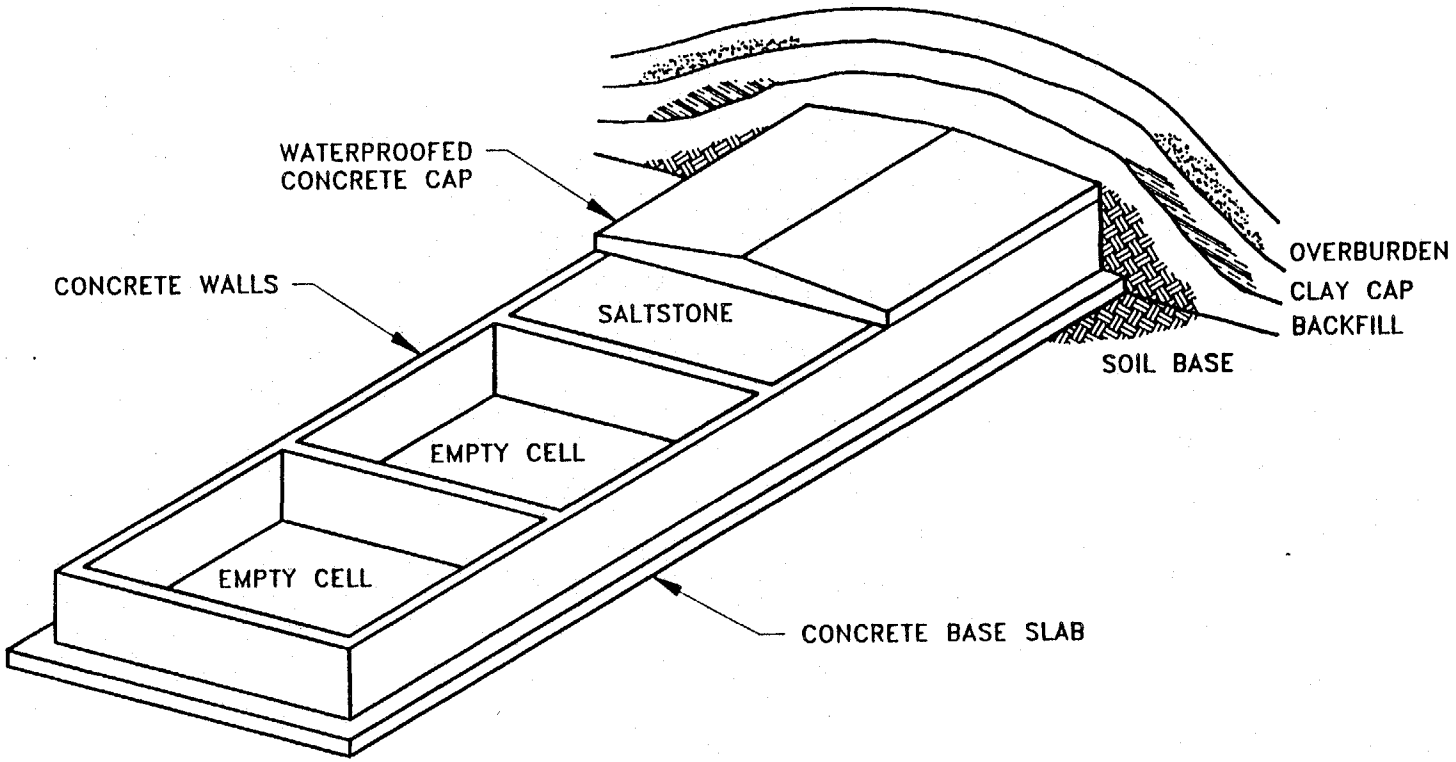
The radioactive waste from plant operations are stored as liquids or slurries in double-walled carbon steel tanks. Tank volume ranges from 750,000-1,300,000 gal. Most of the liquid waste at SRP comes from the two reprocessing plants on site. The separations process is either the Purex or HM process. The Purex process recovers uranium and plutonium from the irradiated fuel. The HM process recovers enriched uranium from uranium-aluminum alloys used as fuel in SRP reactors. Most wastes that comes from these processes are acidic and are neutralized with sodium hydroxide before transfer to mild steel storage tanks. The waste is first transferred to cooled tanks where it remains for about two years until the short-lived fission products have decayed. The waste is then transferred to uncooled tanks. It is probably appropriate to mention at this point that mercury nitrate is used as a catalysis in order to dissolve aluminum with nitric acid. After the waste is neutralized and transferred to tanks, insoluble hydroxides of fission products and of metals that resulted from dissolving the fuel hulls. The volume of sludge is about 10% of the total volume. In recent years, aged supernate has been evaporated and returned to cooled tanks where much of the salt crystallizes. Now the question is, what fraction is high-level waste and what is transuranic and what is low-level waste?

The people at Savannah River have done numerous studies to determine and evaluate potentially useful waste forms. Time does not permit me to discuss all of these in any detail. However, I will just mention calcines, glasses and ceramics, cement and asphalt. A number of studies have been conducted on sludge-supernate separation, fixation of the sludge and cesium separation.

Now I will go into some detail about the saltstone solidification process, which we who worked on the Hanford Grout program have some familiarity.

SALTSTONE

ORNL DWG 93A-85



SALTSTONE SURFACE VAULT, CLAY CAP CONCEPT

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INFORMAL HISTORY OF THE HANFORD GROUT PROGRAM

Earl W. McDaniel

Introduction

The informal history of the Hanford Grout Program (HGP) is based on the author's memory and recollections. Most dates should be considered approximate. A large number of persons from Rockwell Hanford Operations (operating contractor prior to Westinghouse Hanford Company), Westinghouse Hanford Company (WHC), Battelle Pacific Northwest Laboratories (PNL), Kaiser Engineers, and Oak Ridge National Laboratory (ORNL) have made major contributions to the effort to develop, build and operate a Grout Treatment Facility (GTF) at the Hanford site for the permanent disposal of low-level and mixed radioactive waste solutions. In most cases only the facility will be referred to, i.e. PNL, ORNL, WHC, etc., and not individuals.

The Beginning

The present Hanford Grout Program referred to as HGP is the outgrowth of an idea by Joe Wetch, a RHO engineer working on tank waste. Mr. Wetch's involvement with the high-level waste program brought his attention to FUETAP (Formed Under Elevated Temperature and Pressure), which was being developed at ORNL as a candidate waste form for high-level waste.[1]

FUETAP is a tailored cement-based waste form that is cured under elevated temperature to accelerate the hydration (set) of the cement and at elevated pressure to retain the water of hydration

during accelerated cure. After curing, the monolith is dewatered to avoid the possibility of radiolysis. The final product is a highly porous, structurally strong waste form that has demonstrated very low leach rates for Sr, Cs, and Pu.[1]

FUETAP was only half the idea. Mr. Wetch was very interested in ORNL's Hydrofracture Facility (HF) used for the underground disposal of low-level liquid waste solutions.[2,3] The HF involved tailoring a very fluid grout to a specific need and pumping the fluid grout (cement-based materials mixed in proper amounts with the waste solution) into the underlying shale formation that had been "hydrofractured". Mr. Wetch's idea was simply to combine the FUETAP and HF technologies, tailor a waste form to the Hanford defense tank waste, and pump the slurry in to drifts to be drilled into the Basalt Waste Isolation repository being planned for the Hanford site.

Mr. Wetch was successful in obtaining funds through the high-level, lead-site program office, which at that time was at the Savannah River Site. Work started at ORNL on October 1, 1981 (FY-1982). The first effort was devoted to Redox waste. The Redox process predated PUREX for separating U and Pu. Formal reports were never prepared on this early work. A number of other waste streams were evaluated during FY-1982.

Later in the summer, ORNL staff proposed to RL-DOE that instead of pumping waste slurries into BWIP, but that the empty single shell tanks be considered as a repository. This suggestion was based on an informal study made by staff at ORNL and SRL for HQ-DOE. Unfortunately, no report was ever issued on this work.

The proposal was to do an engineering scale demonstration of the technology. After some thought was given to the idea, RL-DOE decided to build the first of a family of grout facilities. It was proposed that a transportable facility be built that could be moved from tank-farm to tank-farm as needed. It should be mentioned that Mr. Wetch went into private industry and was no longer involved in the effort. However, the outgrowth of Mr. Wetch's idea has had a major impact on the waste management efforts at the Hanford Site for the past decade.

Transportable Grout Facility

Work began on a functional design criteria report (FDR) in January 1983. The Transportable Grout Facility(TGF) consisted of a Dry Blend Facility (DBF), the Grout Treatment Facility (GTF), and the disposal trench. The DBF was to be stationary and the GTF to be modular and movable from tank-farm to tank-farm.

The scope of the project was greatly expanded and PNL and Kieser Engineers became part of the Grout Team.

This facility was designed to process very fluid grouts based upon ORNL's hydrofracture facility. The GTF mission was to treat (process) 12-14 million gallons of N-Reactor decontamination solution. The plant was to serve as a pilot plant for two additional plants, to be more robust and to be built in the future.

Sometime in the first year or so, the two additional plants were forgotten in favor of the transportable idea.

Work progressed very rapidly both on the technology and projects side. ORNL was developing formulas to accommodate the

phosphate/sulfate reactor decontamination solution, which was a high Ph sodium phosphate/sodium sulfate solution not regulated and containing little radioactivity. PNL's role was that of formula verification and variability studies to assure that work done at ORNL was valid and that it had an acceptable range of variability.[4] Formulation efforts were highly successful for the phosphate/sulfate waste. The formulation effort was then expanded to include cladding removal waste and double-shell tank waste (DST). These topics will be addressed in a separate section.

Plant Design

Design and construction of the Dry Solids Blending Facility went well. This section was completed according to schedule. However, the A/E firm awarded the contract to build the GTF had major problems with costs associated with remote maintenance and the firm defaulted on the contract. Several years were lost due to this default.

There were several events during this time frame that had a major affect on the Grout Program: (1) Westinghouse Hanford Company (WHC) replaced RHO as the site operating contractor and (2) the N-reactor was put on standby and finally closed completely. With the closing of the N-reactor the source of phosphate/sulfate waste was eliminated.

It was becoming apparent to a number of site personal that a more robust plant should be built. However, since the Dry Solids Blending Facility was sized to support the TGF, the decision was made to keep it the same size.

It should be pointed out that the N-reactor did not close until well into construction of the plant (TGF).

Cladding Removal Waste

A significant amount of development work was performed on cladding removal waste at both PNL and ORNL. Successful formulas were developed to process this waste. Analysis of the waste determined it to be TRU waste and not a suitable candidate for grouting. All development work was stopped on this waste stream.

Work focused on doing an operational campaign with the existing phosphate/sulfate waste as soon as the plant was completed. In the mean time, process development continued on DST. During this transitional period, shallow land disposal trenches were replaced with below grade RCRA vaults.

Each subsurface vault was to be constructed of reinforced concrete. Inside dimensions of the vaults are approximately 125 feet long, 50 feet wide, and 34 feet deep. In compliance with Washington State regulations, liners and liquid collection systems will be installed beneath each vault.

Each vault will hold about 1.4 million gallons of grout. Asphalt-based barriers are built around the vault to minimize the diffusion of radionuclides to the environment and to satisfy long-term environmental protection requirements.

Summary Description of Hanford Grout Facility

The Grout Facility consists of a Dry Materials Facility, the Grout Processing Facility, and the Grout Disposal Facility (near surface vaults).

The Dry Materials Facility receives, stores, and blends commercially produced cement-based or natural materials that will later be mixed with liquid waste to produce a grout.

Examples of these dry materials are portland cement, fly ash, blast furnace slag, and clays. After the materials are blended, they are hauled to the Grout Processing Facility in 25-ton capacity transport trucks.

The Grout Processing Facility mixes the blended solids with the low-level liquid waste to form a grout. This facility was designed to pump the resulting mixture through a pipeline to the disposal vault at a rate of 50-60 gallons per minute.

Each concrete vault will hold about 1.4 million gallons of grout. The vaults are built in compliance with the State of Washington regulations. Liners and liquid collection systems are installed beneath each vault. The systems will contain and collect any excess water, especially during the initial period while the grout is hardening. The vault's inside dimensions are approximately 125 feet long, 50 feet wide, and 34 feet deep.

Summary Project History

The Dry Materials Facility was completed in 1986. The Grout Processing Facility and the first vault were completed in 1988. A demonstration run, resulting in grouting a million gallons of non-hazardous (phosphate/sulfate) low-level liquid waste, was completed in July 1989. Four additional vaults have been completed (1992) for proposed future grouting campaigns.

HISTORICAL REVIEW OF USDOE TANK WASTE MANAGEMENT

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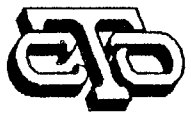
A Short Course Presented for
Westinghouse Hanford Company
February 15-16, 1995



OBJECTIVE

The objective of this two-day short course is to present historical overview of the DOE tank waste activities. Material covered is readily available in the open literature. There is no intent to be critical of past practices. As I proceed through the course I will inject world events that impacted the United States nuclear program or, in some cases, how world events were impacted by the United Sates nuclear program.

As the audience is well aware, both solid and liquid waste results from most nuclear operations. In my discussion I will confine myself to liquid, mostly tank waste, and sludge. We will start out at the dawn of the nuclear age and end up at the Hanford site in 1995.

CHRONOLOGY

1895	Wilhelm Roentgen discovers X-ray radiation.
1932	James Chadwick discovers the neutron.
1938	Otto Hahn and others achieve atomic fission.
October 1939	Albert Einstein and others persuade FDR to support research on an atomic bomb.
December 1941	FDR approves the Manhattan Project.
December 1942	Enrico Fermi and others achieve controlled and sustained chain reaction.
July 1945	First atomic bomb detonated at Alamogordo, New Mexico.
August 1945	Hiroshima and Nagasaki are destroyed by atomic bombs.
June 1946	U.S.A. proposes Baruch Plan for international control of nuclear weapons; U.S.S.R. rejects it.
June 1946	First Atomic Energy Act enacted.
August 1949	U.S.S.R. detonates its first fission bomb.

CHRONOLOGY (CONTINUED)

January 1950	President Truman approves development of the hydrogen bomb.
May 1950	NSC-68 outlines plan for massive U.S. rearmament and military spending.
January 1951	AEC begins weapons testing at Nevada test site.
December 1951	First experimental production of electricity from a nuclear reactor.
November 1952	U.S.A. detonates its first hydrogen bomb, followed by U.S.S.R. in 1953.
December 1953	President Eisenhower's Atoms for Peace address.
January 1954	John Foster Dulles announces a new strategic policy, massive retaliation.
June 1954	AEC affirms denial of security clearance to J. Robert Oppenheimer.
August 1954	Second Atomic Energy Act enacted.
September 1957	Congress enacts the Price-Anderson Act, indemnifying the nuclear power industry in the event of nuclear accidents.

CHRONOLOGY (CONTINUED)

October 1957	U.S.S.R. launches Sputnik. The missile gap is born.
December 1957	Shippingport, the world's first exclusively non-military power reactor, comes on stream.
? 1957	U.S.S.R. nuclear disaster near Sverdlovsk.
Spring 1958	Worldwide protests begin against atmospheric nuclear testing.
January 1961	President Kennedy advances a new strategy, flexible response.
June 1962	Robert McNamara announces No Cities doctrine.
July 1963	U.S.A., U.S.S.R., U.K. sign Partial Test Ban Treaty, ending atmospheric testing.
February 1964	Mutually assured destruction becomes the nuclear strategy of U.S.A.
October 1966	Nuclear accident at Fermi-1, a commercial breeder reactor near Detroit.
September 1967	U.S.A. announces it will deploy a "thin" antiballistic missile system.

CHRONOLOGY (CONTINUED)

January 1972	AEC generic rule-making hearings begin on ECCS.
May 1972	SALT I is signed in Moscow. ABM deployment is sharply limited.
August 1974	Rasumussen report released, downplaying likelihood of reactor accidents.
October 1974	Congress abolishes AEC, substituting NRC and ERDA (which is later replaced by Department of Energy).
November 1974	Karen Silkwood killed in accident.
November 1974	U.S.A., U.S.S.R. agree to Vladivostok accords
January 1975	Browns Ferry reactor accident.
March 1979	Three Mile Island; concurrent, but coincidental release of film, *The China Syndrome.*
June 1979	SALT II signed in Moscow.
December 1979	SALT II withdrawn from Senate by President Carter.

A SELECTION OF WORLD EVENTS THAT IMPACTED NUCLEAR WEAPONS DEVELOPMENT

- Potsdam Conference, July, 1945
- Atomic bombing of Japan, August, 1945
- David Greenglass, a machinist at LASL, works on implosion lens of Pu bomb.
- Russians block surface access to Berlin, 1948—49
- Russia explodes first nuclear device, Aug., 1949
- Korean conflict starts June, 1950
- Savannah River Site starts producing nuclear materials, 1953

A SELECTION OF WORLD EVENTS THAT IMPACTED NUCLEAR WEAPONS DEVELOPMENT (CONTINUED)

- Gaseous diffusion facilities expanded
- Additional reactors built at Hanford
- Rocky Flats Plant built, Dow becomes operator 1953
- McCarthy with hunt starts, 1950
- Julius and Ethyl Rosenberg, atomic spies electrocuted June, 1953
- Birth of Nuclear Navy, 1950
- Light water reactors commercialized, 1956
- Atoms for peace program, 1953

A SELECTION OF WORLD EVENTS THAT IMPACTED NUCLEAR WEAPONS DEVELOPMENT (CONTINUED)

- John Foster Dulles offers French nuclear weapons to use in Indo-China, 1954
- Camp Century in Greenland built, 1960
- U-2 incident, 1960
- John F. Kennedy assassinated, 1963
- China explodes nuclear device, 1964
- USSR breakup, 1991

MAJOR WORLD WAR II SITES

The major Manhattan Project sites of World War II were:

- The Hanford Engineering Works, Richland, WA
- Clinton Engineering Works, Oak Ridge, TN
- Los Alamos Scientific Laboratory, Los Alamos, NM
- Columbia University, Upton, LI, NY
- University of Chicago Metallurgical Lab., Chicago, IL

MAJOR WORLD WAR II SITES (CONTINUED)

The other sites did not generate fuel processing wastes. Sites that were built in the 1950/60s that will be discussed are:

- Savannah River Site, Akin, SC
- National Reactor Test Center, Idaho Falls, ID
- West Valley Nuclear Services, West Valley, NY
- Rocky Flats Plant, Golden, CO

BACKGROUND FOR DOE DECISIONS FOR THE DISPOSITION OF THE HLW

After the West Valley Demonstration Project (WVDP) started, the Department of Energy had to decide how they wanted the waste to be handled. There were several alternatives considered as approaches to solidify the HLW into a form suitable for disposal in a federal repository.

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  - Separated salt\sludge process-remaining high-level radioactive components after decontamination of supernatant mixed with HLW sludge
  - Nonseperated salt\sludge—no separation or concentration

BACKGROUND FOR DOE DECISIONS FOR THE DISPOSITION OF THE HLW (CONTINUED)

- On-site processing to an Interim Waste Form/Liquid HLW converted to solid interim waste form for future processing to terminal form
- In-tank solidification/HLW mixed with cement and poured into existing tanks
- No action

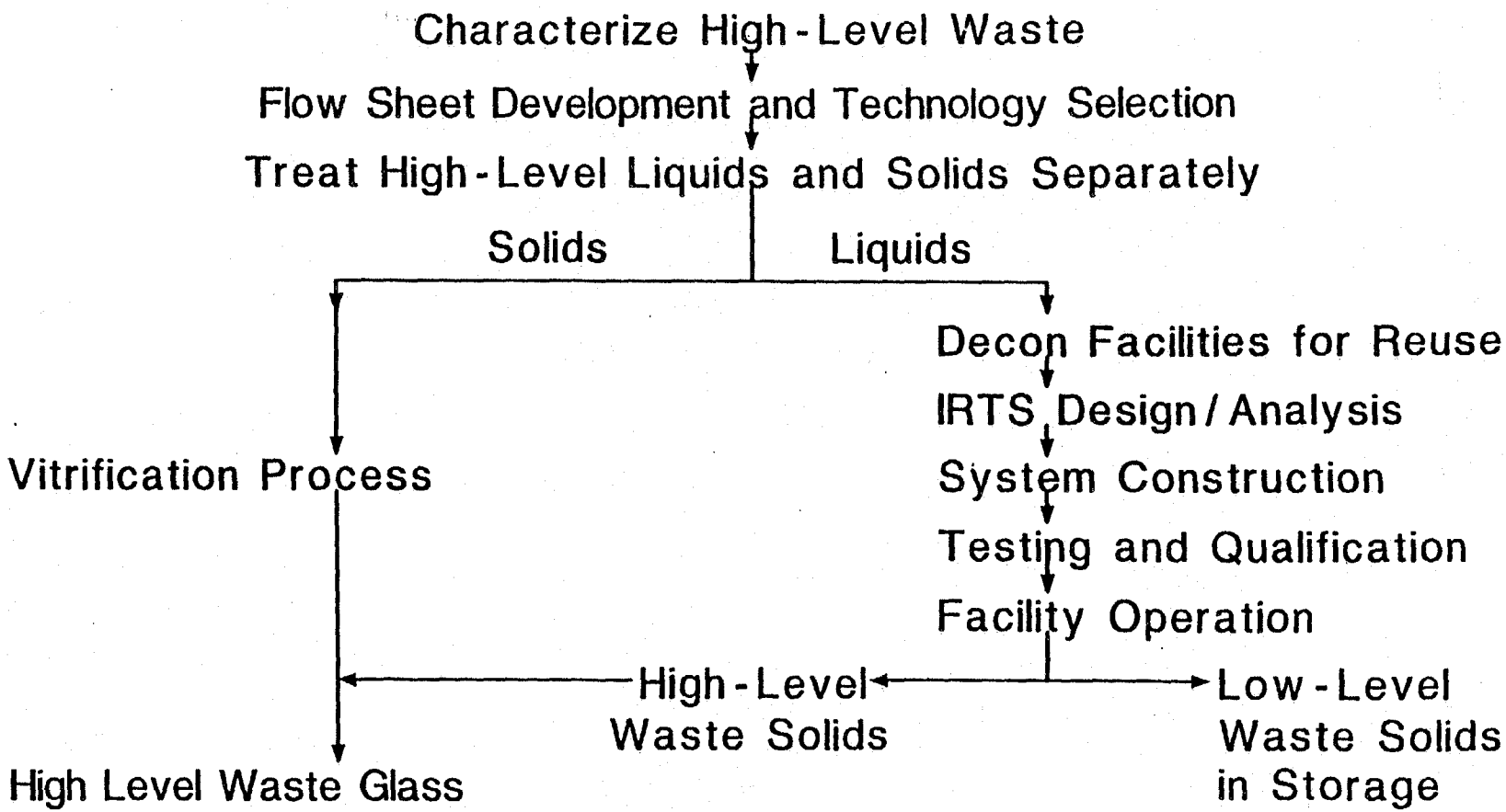


Figure 1-1
High-Level Waste Processing Logic Diagram

PROCESS OVERVIEW

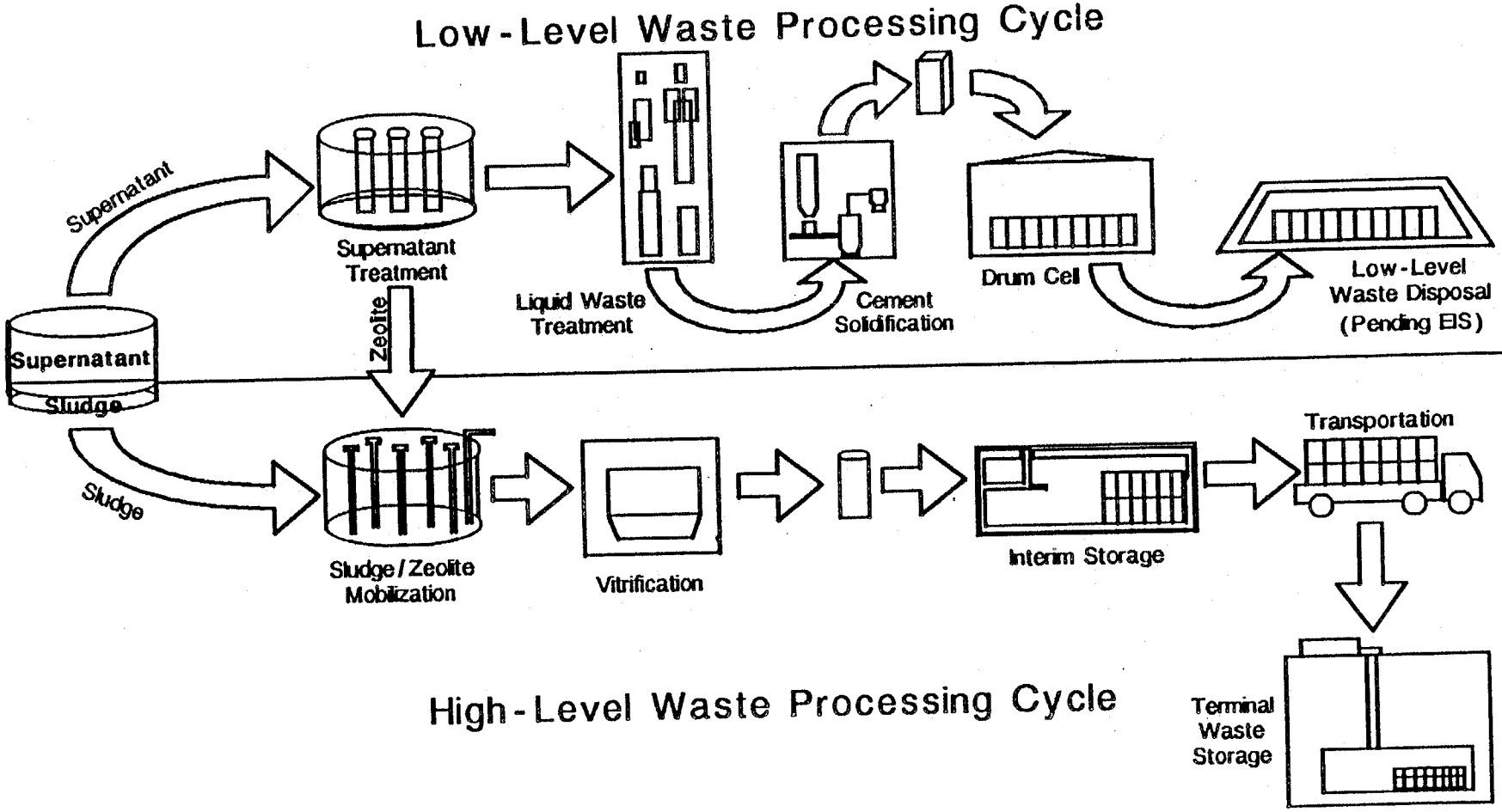


Figure 1-2
Conceptualized High-Level Process Flow Diagram

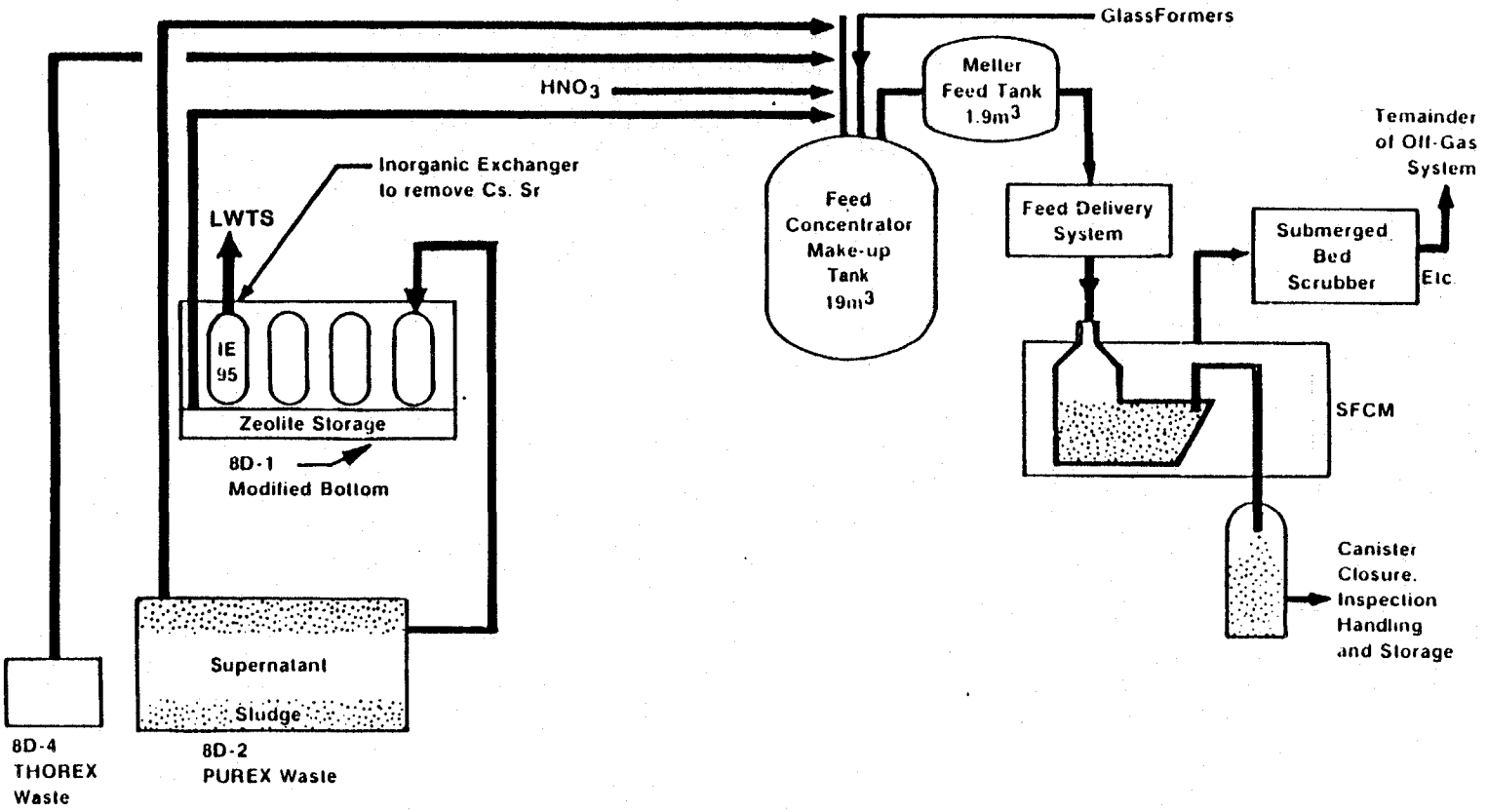


FIGURE 1
HLW Processing Flow Sheet

SYSTEMS BUILT TO TREAT HLW AND LLW AT WVDP

- CEMENTATION SOLIDIFICATION SYSTEM (CSS)
- LIQUID WASTE TREATMENT SYSTEM (LWTS)
- SUPERNATANT TREATMENT SYSTEM(STS)
- WASTE IMMOBILIZATION SYSTEM (WMS)
- VITRIFICATION SYSTEM (VS)
- HIGH LEVEL WASTE INTERIM STORAGE
- SIZE REDUCTION FACILITY
- TRANSURANIC ASSAY AND INTERIM STORAGE

PRECIPITATION METHODS CONSIDERED

- Ferrocyanide
- PTA
- NaTBP

The highest decon factors were obtained using IE-95/96. It is worth noting that IE-95 was successfully used at Hanford in the 1950s to remove Cs from PUREX supernatant. In addition to the West Valley facility, reprocessing plants were built by the following:

TABLE 1-1: 8D-2 SUPERNATANT CHEMICAL COMPOSITION

Compound	Wt. % Wet Basis	Wt. % Dry Basis	Total Kg in Supernatant
$NaNO_3$	21.10	53.38	602,659
$NaNO_2$	10.90	27.57	311,326
Na_2SO_4	2.67	6.76	76,261
$NaHCO_3$	1.49	3.77	42,557
KNO_3	1.27	3.21	36,274
Na_2CO_3	0.884	2.24	25,249
NaOH	0.614	1.55	17,537
K_2CrO_4	0.179	0.45	5,113
NaCl	0.164	0.42	4,684
Na_3PO_4	0.133	0.34	3,799
Na_2MoO_4	0.0242	0.06	691
Na_3BO_3	0.0209	0.05	597
$CsNO_3$	0.0187	0.05	534
NaF	0.0176	0.04	503
$Sn(NO_3)_4$	0.00859	0.02	245
$Na_2U_2O_7$	0.00808	0.02	231
$Si(NO_3)_4$	0.00806	0.02	230
$NaTcO_4$	0.00620	0.02	177
$RbNO_3$	0.00416	0.01	119
Na_2TeO_4	0.00287	0.007	82
AlF_3	0.00271	0.007	77
$Fe(NO_3)_3$	0.00152	0.004	43
Na_2SeO_4	0.00054	0.001	15
$LiNO_3$	0.00048	0.001	14
H_2CO_3	0.00032	0.0008	9
$Cu(NO_3)_2$	0.00022	0.0005	6
$Sr(NO_3)_2$	0.00013	0.0004	4
$Mg(NO_3)_2$	0.00008	0.0002	2
TOTAL	39.53	100.00	1,129,038
H_2O (by difference)	60.47		1,727,164

TABLE 1-2: RADIOCHEMICAL COMPOSITION OF 8D-2 SUPERNATANT

Species	1 ft*	5 ft	15 ft	% RSD**
	mCi/gm			
Cs-137	2.86 E0	2.80 E0	2.84 E0	0.2
Cs-134	2.36-E2	2.32-E2	2.35-E2	3.0
Sr-90	1.14-E3	1.13-E3	1.12-E3	1.0
Sb-125	5.7-E5	5.5-E5	5.4-E5	12.0
Ru-106	< 1.5-E5	< 1.5-E5	< 1.5-E5	NA
Ce-144	< 7.6-E7	< 7.6-E7	< 7.6-E7	NA
Rare Earth B	2.1-E4	1.5-E4	2.7-E4	3.0
Am-241	< 1.5-E5	< 1.5-E7	< 1.5-E7	NA
Am-243	< 2-37	< 2-E7	< 2-E7	NA
Cm-244	< 6-E8	< 6-E8	< 6-E8	NA
	μgm/gm			
Pu-238	0.0024	0.0031	0.0027	3.0
Pu-239	0.1302	0.1545	0.1461	3.0
Pu-240	0.0251	0.0307	0.0301	3.0
Pu-241	0.0051	0.0066	0.0060	3.0
Pu-242	0.0021	0.0025	0.0024	3.0
U-233	0.017	0.019	0.018	8.0
U-234	0.014	0.017	0.016	8.0
U-235	0.967	1.101	1.044	3.0
U-236	0.097	0.108	0.105	8.0
U-238	55.145	63.182	59.865	0.3

*Samples taken at three levels: 1, 5, and 15 feet below the surface of the liquids.

** Percent relative standard deviation.

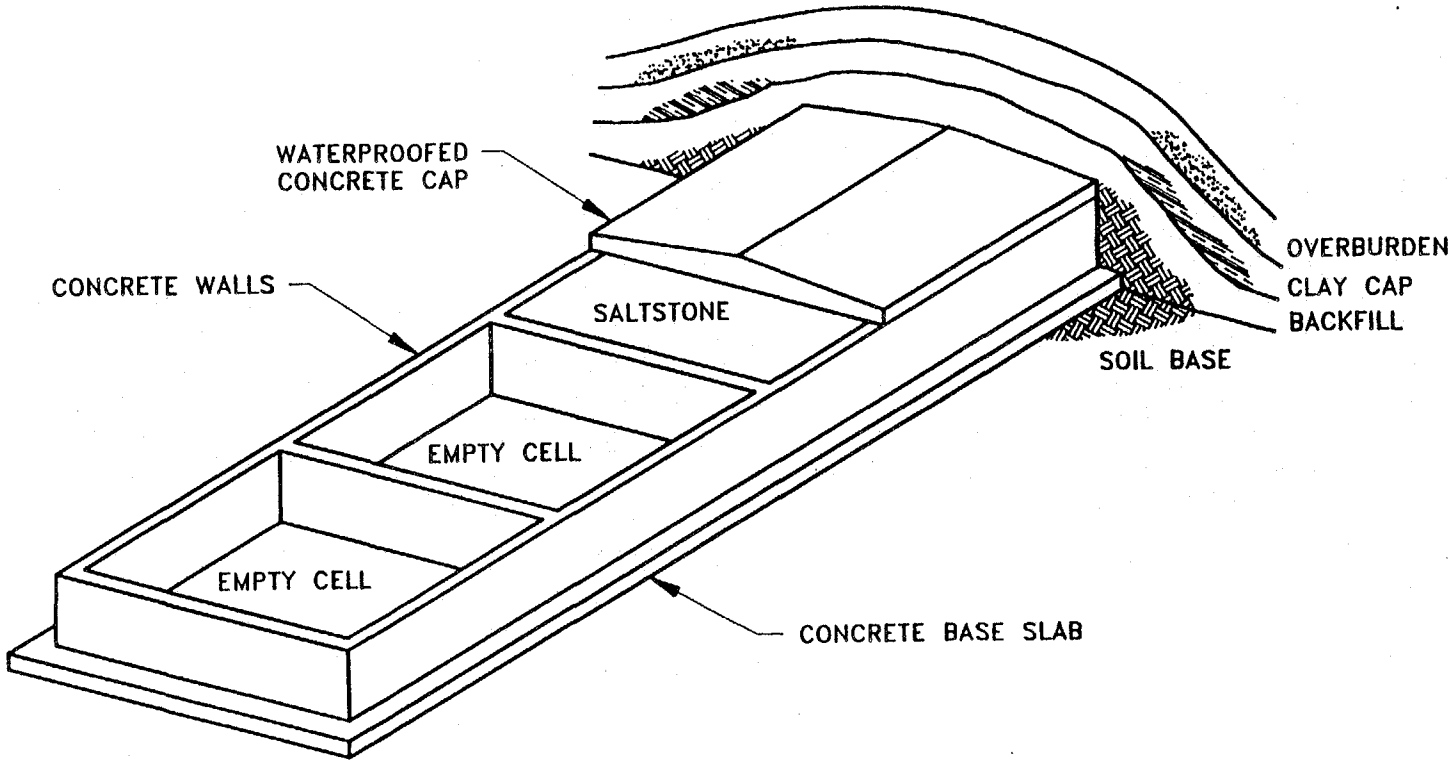
OTHER REPROCESSING SITES

In addition to the West Valley facility, other reprocessing plants planned or built were:

- General Electric's Midwest Fuels Reprocessing Plant at Morrie, Ill.
- Allied General Nuclear Services, at Barnwell, SC
- A plant to be operated by Exxon Nuclear Corp. was scheduled to be built in Tennessee.

None of the above plants ever operated.

ORNL DWG 93A-85



SALTSTONE SURFACE VAULT, CLAY CAP CONCEPT

CLINTON ENGINEERING WORKS WAS DIVIDED INTO THREE DISTINCT ACTIVITIES

- Clinton Laboratories, now Oak Ridge Laboratory, at one time Hollifield National Laboratory coded, the X-10 site.
- Y-12 Electromagnetic Plant, this plant was coded Y-12.
- The Gaseous Diffusion Plant, coded K-25.

HANFORD GROUT PROGRAM
A HISTORICAL REVIEW
1981 -1991

EARL W. MCDANIEL

CHEMICAL TECHNOLOGY DIVISION
OAK RIDGE NATIONAL LABORATORY
OAK RIDGE, TENNESSEE 37831

- FY 1982
  - Grout development on Redox sludge supported by HLW Program

- FY 1983
  - Demonstration facility
  - During 1st quarter decision made to build operating facility
  - Facility to be 1st of 3
  - Facility dedicated to nuclear reactor decontamination waste
  - Expect 12-14 1M gal campaigns

- FY 1984-1985
  - Decision to make plant transportable and build only one plant
- FY 1985
  - Contract awarded to ATCOR
  - ATCOR defaulted on contract
- FY 1986
  - Contract awarded to ATI
  - Development started on DSSF
- FY 1989
  - Successful completion of one million gal campaign - July 1989
  - All development work reported in documents cleared for public release

- Oak Ridge National Laboratory
  - Formulation/Process Development
- Battelle-Pacific Northwest Laboratories
  - Verification
  - Variability study
  - Leach testing
- Westinghouse-Hanford Company
  - Customer
  - Hot cell studies
  - Waste characterization

DEVELOPMENT STEPS

- Laboratory
  - ORNL
  - PNL
  - WHC
- Pilot Scale
  - PNL
- Cold Tests & Operator Training
  - WHC

ALL WORK WAS PERFORMED TO

N-QA-1 SPECIFICATIONS

DEVELOPMENT OF DSSF FORMULA

- WHC believed SRS was leader in grout development
- ORNL instructed to base development on that of SRS
- Pilot scale determined heat to be a major problem

Grout Properties of Interest

	Preferred Value
Critical flow rate	< 60 GPM
10 min. gel strength	< 100 $lb_f/100\ ft^2$
28-d freestanding liquid	< 0.5 vol %
28-d unconfined compressive strength	> 500 psi
Nitrate and Nitrite Leachability Index	> 6
Heat Evolution	< 90° C

ORNL

WASTE IMMOBILIZATION ARCHIVES

DOUBLE SHELL SLURRY FEED (DSSF)

(1986-1990)

Data Acquisition Conditions

ORNL DSSF Archives

- Standardized Procedures, ASTM, API, ANS
- Strict Quality Assurance
- Detailed Documentation

Archives Data Review

(DSSF)

- Dry Solid Blends Tested
  >40
- Dry Solid Blend Components Tested
  >15
- Waste Dilutions Tested
  5
- Processability Data Entries
  >8,000
- Leachability Data Entries (ANS-16.1)
  >16,000

FY 1991-92
DEVELOPMENT WORK

Developing New 106-AN Grout Formulation

Potential ingredients tested

- Type II-LA (moderate heat of hydration) Portland cement
- Ground granulated blast furnace slag (GGBFS)
- Type V Portland cement
- Class H cement
- Class F fly ash
- Ground limestone

Developing New 106-AN Grout Formulation (continued)

- Ground air-cooled blast furnace slag (GABFS)
- Air-cooled blast furnace slag aggregate
- Natural, rounded sand
- Attapulgite clay
- Sodium silicate powder
- Slaked lime
- 6 admixtures (2 fluidizers, 2 set modifiers, 2 air entrainers)

Selected Ingredients

	Mix ratio variable lb/gal	Preliminary recommended formulation lb/gal
Type II-LA Portland cement	W_1	2.5
Class F fly ash	W_2	1.2
Attapulgite clay	W_3	0.8
GABFS	W_4	3.5
Total		8.0

MIXTURE EXPERIMENT

METHOD OF DETERMINING THE EFFECT OF THE GROUT COMPOSITION ON THE GROUT PROPERTIES WITH THE MINIMUM NUMBER OF EXPERIMENTS

Mixture Experiment Design With Following Bounds Centered Around Preliminary Recommended Formulation

Bounds (Units of lb ingredient/gal 106-AN)

$$1.5 \leq W_1 \leq 3.5$$
$$0.0 \leq W_2 \leq 6$$
$$0.5 \leq W_3 \leq 2$$
$$0.0 \leq W_4 \leq 6$$
$$4.0 \leq W_2 + W_4 \leq 6$$
$$W_1 + W_2 + W_3 + W_4 \leq 10$$

FUETAP

(FORMED UNDER ELEVATED TEMPERATURE AND PRESSURE)

EARL W. MCDANIEL
CHEMICAL TECHNOLOGY DIVISION
OAK RIDGE NATIONAL LABORATORY

FUETAP WAS DEVELOPED AT
OAK RIDGE NATIONAL LABORATORY
DURING 1977 - 81 AS A CANDIDATE
HIGH-LEVEL WASTE FORM.

IN 1981 BOROSILICATE GLASS WAS CHOSEN AS THE WASTE FORM FOR HIGH-LEVEL WASTE.

DEVELOPMENT ON FUETAP TERMINATED AT THE END OF FY 1981.

DEVELOPMENT RESULTED IN ONE 3/5 SCALE ENGINEERING EXPERIMENT AFTER EXTENSIVE LABORATORY STUDIES.

CANDIDATE WASTE FORM PROCESSES EVALUATED FOR IMMOBILIZATION OF COMMERCIAL HIGH-LEVEL WASTES

In-Can Glass Melter

Joule-Heated Glass Melter

Glass Ceramic

Marbles-in-Lead

Supercalcine Pellets-in-Metal

Carbon-Coated Pellets-in-Metal

Supercalcine, Hot-Isostatic Pressed

SYNROC, Hot-Isostatic Pressed

Titanate

Concrete

Cermet

FINAL PRODUCT, PROCESS, AND COMBINED FIGURE-OF-MERIT SCORES FOR THE SEVEN CANDIDATE WASTE FORMS

WASTE FORM	PRODUCT	PROCESS	COMBINED[a]
Borosilicate Glass	67	83	75
SYNROC	95	42	63
Tailored Ceramic	93	42	62
High-Silica Glass	64	51	57
FUETAP Concrete	39	77	55
Coated Particles	87	32	53
Glass Marbles in a Lead Matrix	40	58	48

[a]Geometric mean

QUESTIONS

- WHAT IS FUETAP'S
  - — FAVORABLE PROPERTIES
  - — UNFAVORABLE PROPERTIES
- FLOW SHEET
- APPLICATIONS

- FUETAP IS A TAILORED INORGANIC MINERAL-BASED WASTE FORM
- PORTLAND CEMENT IS USED AS A BINDER
- IT IS SIMILAR TO A MORTAR
- IT IS NOT CONCRETE

FAVORABLE PROPERTIES

- LOW TEMPERATURE PROCESS
- USES LOCALLY AVAILABLE MATERIALS
- REQUIRED PROCESS EQUIPMENT IS STATE-OF-THE-ART.
- WASTE FEED CAN BE SOLID, LIQUID, OR SLUDGE
- HIGH STRUCTURAL STRENGTH

FAVORABLE PROPERTIES (continued)

- LOW LEACH RATES FOR Sr, Cs, AND Pu
- DEWATERING ELIMINATES POTENTIAL FOR RADIOLYSIS
- RADIOACTIVE ELEMENTS IN MANY CASES ARE INCORPORATED INTO THE MINERAL STRUCTURE

UNFAVORABLE PROPERTIES

- HIGH WASTE LOADING (>50 WT. %) COULD RESULT IN LARGE VOLUME INCREASE
- WASTE FORM IS VERY POROUS
- MICROFRACTURES DEVELOP AS DEWATERED WASTE FORM COOLS

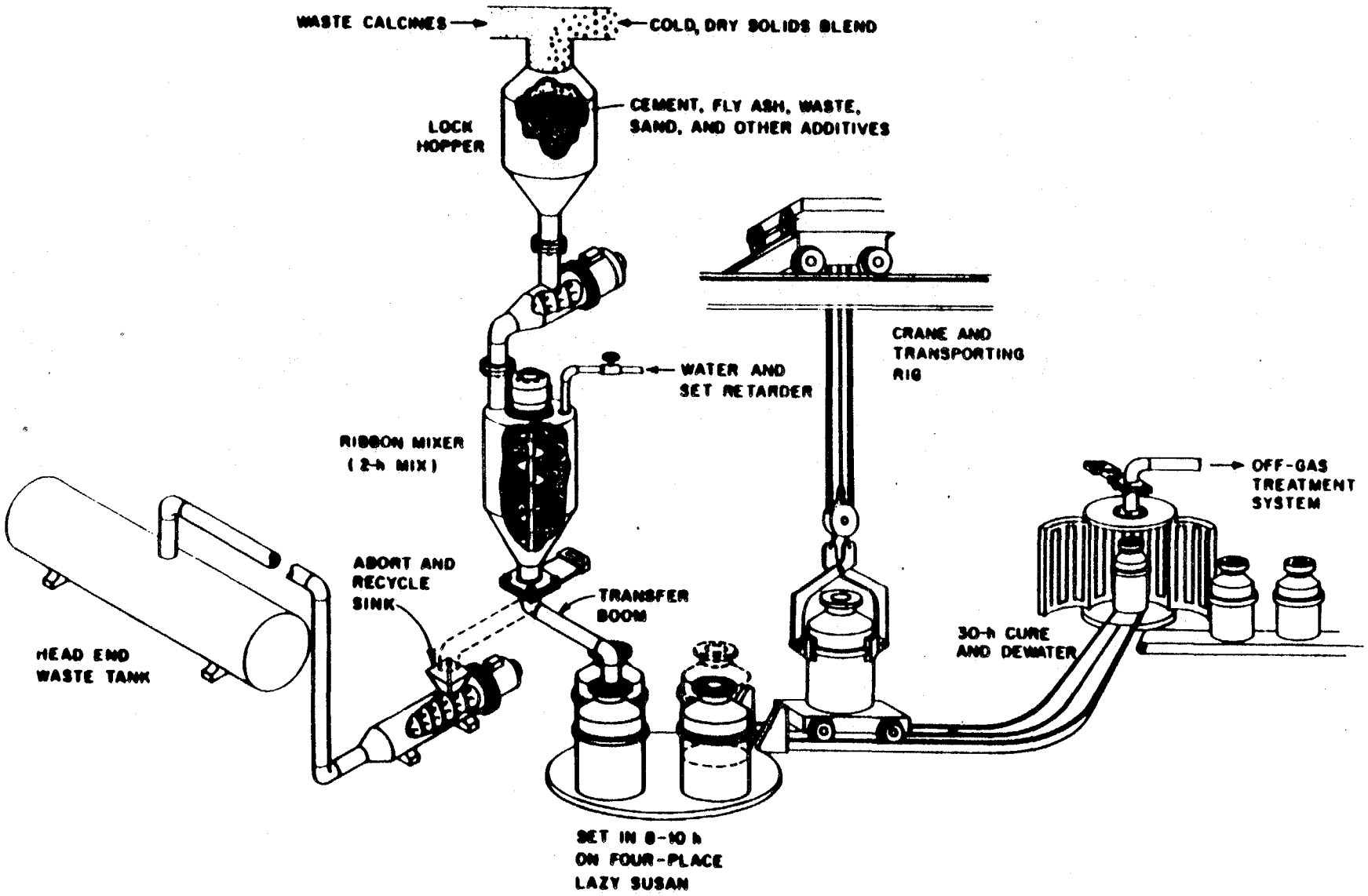

WASTE CALCINES
COLD, DRY SOLIDS BLEND
LOCK HOPPER
CEMENT, FLY ASH, WASTE, SAND, AND OTHER ADDITIVES
CRANE AND TRANSPORTING RIG
WATER AND SET RETARDER
RIBBON MIXER (2-h MIX)
OFF-GAS TREATMENT SYSTEM
ABORT AND RECYCLE SINK
TRANSFER BOOM
HEAD END WASTE TANK
30-h CURE AND DEWATER
SET IN 8-10 h ON FOUR-PLACE LAZY SUSAN

Table 3. FUETAP concrete mix compositions

Component	Mix 1 (wt %)	Mix 2 (wt %)
Cement	37.5	38.9
Fly ash	8.9	9.3
Indian red clay	8.9	9.3
Simulated sludge	14.4	14.8
Suspending agent	0.4	0.4
$NaNO_3$	2.6	0.0
Water	27.3	27.3

Table 4. Simulated waste sludge

Oxide	wt %
Fe_2O_3	75.18
Al_2O_3	14.89
Cr_2O_3	4.94
NiO	4.78
SrO	0.14
$Ru(OH)_3$	0.03
CeO_2	0.04

Table 5. Properties of initial FUETAP concretes[a]

Cement type	Mix[b] No.	Compressive strength[c] (MPa)	Porosity[d] (%)	Density (g/cm^3)	Thermal conductivity[e] (W/mK)	Water loss[f] at 250°C (%)
1	1	14.2	47.6	1.56	0.44	46.9
	2	13.1	51.4	1.54	0.46	12.1
3	1	14.0	46.6	1.54	0.46	62.6
	2	14.9	47.2	1.52	0.43	17.7
HAC[g]	1	h	31.0	i	i	72.2
	2	9.4	49.8	1.40	0.53	14.5

[a]Solidified and cured 24 hr at 250°C and 600 psi.

[b]See Table 3 for composition. Mix 1 contains 2.6 wt % $NaNO_3$, the equivalent of 1 M nitrate in the sludge. Mix 2 contains no nitrate.

[c]Pressure required to crush a right-circular cylinder 4.8 cm diam by 10.2 cm.

[d]Mercury porosimetry measurements at 15,000 psi.

[e]Thermal conductivity at 103°C.

[f]Total initial water lost when specimens were heated at 250°C to constant weight; total heating time, 24 hr.

[g]Lumnite, a high-alumina cement (HAC), for analysis of cements and fly ash (see Table 6).

[h]Specimen was faulty and could not be tested.

[i]Not available.

TAILORING AN INORGANIC MINERAL-BASED WASTE FORM

- IDENTIFY SPECIFIC REQUIREMENTS
- IDENTIFY TYPES AND SOURCES OF MATERIALS
- DEVELOP WASTE FORM QUALIFICATION STRATEGY

SPECIFIC REQUIREMENTS

- BASED ON WASTE TYPE i.e., TRU, TRU/MIXED etc.
- VOLUME OF WASTE
- PHYSICAL PROPERTIES OF WASTE
- CHEMICAL PROPERTIES OF WASTE
- APPLICABLE REGULATIONS, DOE, STATE, NRC, EPA, OTHER

INORGANIC MINERAL-BASED MATERIALS

- FLY ASH
- BLAST FURNACE SLAG
- SAND
- ZELOLITES
- CLAYS
- SILICA FUME
- CEMENT

WASTE FORM QUALIFICATION STRATEGY

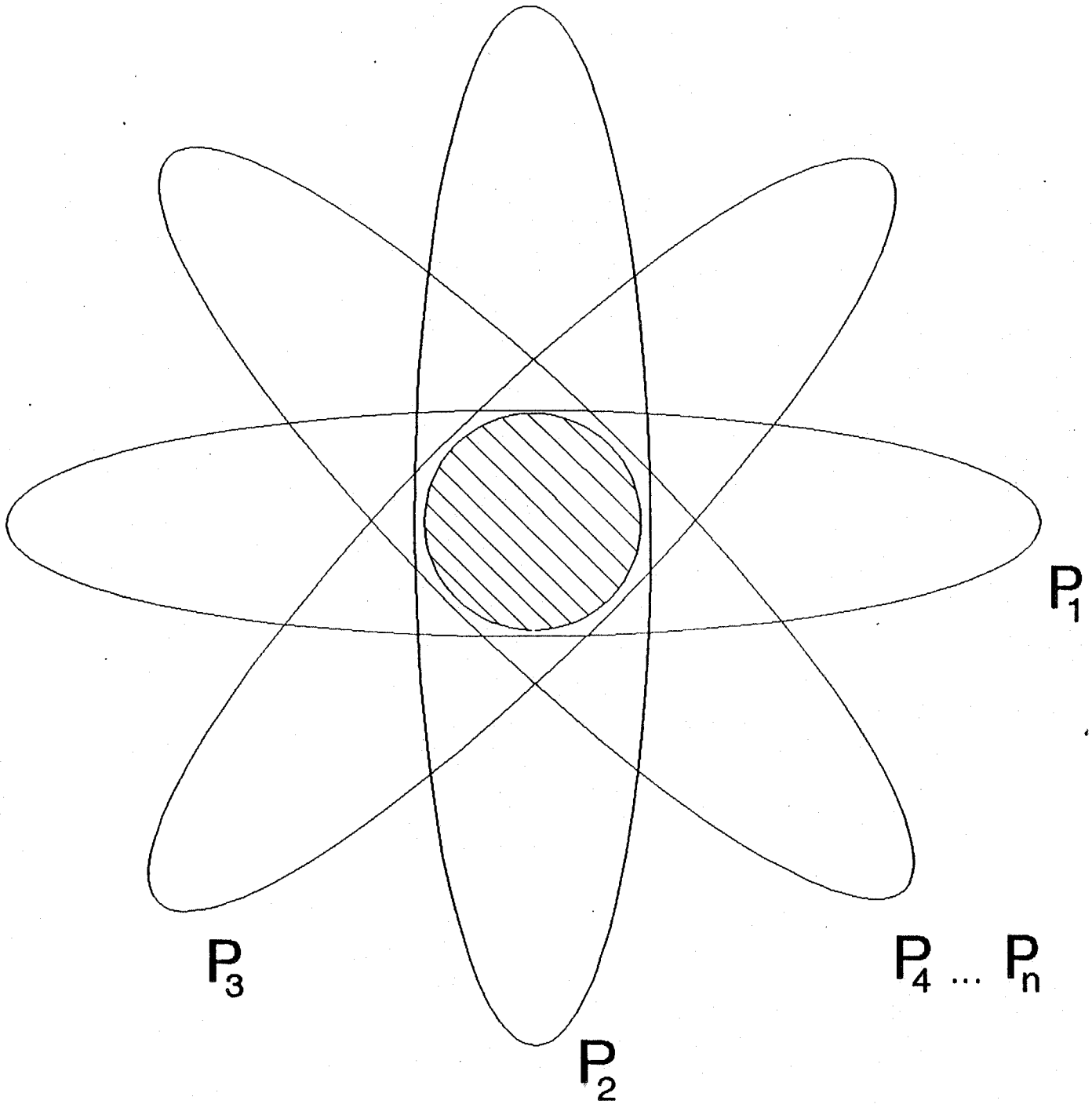
STEPS IN WASTE FORM QUALIFICATION (WFQ)

- DEVELOP ACCEPTANCE CRITERIA
- WASTE FEED CHARACTERIZATION
- SELECTION OF TREATMENT TECHNOLOGIES (SOLIDIFICATION
- SCOUTING TESTS TO DETERMINE IF CHOSEN TECHNOLOGIES ARE APPLICABLE

STEPS IN WASTE FORM QUALIFICATION (WFQ) (continued)

- DESIGN STATISTICALLY MEANINGFUL EXPERIMENTS TO SUPPORT SELECTION
- DATA ANALYSIS AND DETERMINATION OF LIMITS OF ACCEPTABILITY
- PILOT-SCALE TESTS
- FULL-SCALE DEMONSTRATION

OPERATING WINDOW



A WASTE FORM IS NOT FULLY QUALIFIED UNTIL ALL SUPPORT DOCUMENTS HAVE BEEN REVIEWED BY THE APPROPRIATE AGENCIES AND ACCEPTED.

APPROVAL IS THEN GRANTED FOR PLANT START-UP.

ALL WORK IS DONE IN ACCORDANCE TO N-QA-1 REQUIREMENTS.