OVERVIEW OF HIGH-LEVEL WASTE MANAGEMENT ACCOMPLISHMENTS

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INTRODUCTION

Processing of fuels from defense reactors generated the first high-level nuclear wastes during the mid 1940's. The liquid wastes were placed in shielded carbon steel tanks. It was always assumed that some satisfactory method would be developed to dispose of these wastes. As the nuclear power industry progressed, further thoughts developed to handle the liquid wastes and to provide final disposal. The moratorium in commercial nuclear fuel reprocessing has complicated the issues of waste management and disposal. In one way, however, this pause in fuel reprocessing has assisted in alleviating a potential constraint of waste disposal by providing radioactive decay time which reduces the heat load of waste forms.

Early in the storage of liquid wastes, single walled tanks backed up by soil sorption was used. Double walled tanks were later constructed to provide better containment. Because liquids can be difficult to control in the event of tank leakage, researchers contemplated solidification of the waste to reduce dispersability. Many solidification concepts were investigated to provide additional assurance of control. To culminate appropriately safe disposal of high-level wastes, the concept of stable geological repositories was developed.

The breadth of studies and operations are now extensive. Laboratory tests, pilot-plant operations, full-scale operating plants and geological disposal evaluations are examples of these efforts. Federal agencies, such as the Nuclear Regulatory Commission and the Environmental Protection Agency, are developing criteria for radioactive waste disposal.

LIQUID STORAGE

High-level liquid waste storage at Hanford has been in single

shell carbon steel tanks.[1] Leaks from these tanks to the ground have resulted in replacement of the old tanks with double walled carbon steel tanks; the replacement is planned for completion by the end of 1981. Leaks in the old tanks appear to be caused by stress corrosion; the new tanks are annealed to avoid this problem. The lifetime of the new double shell tanks is estimated to be at least 50 years.

At the Savannah River Plant (SRP), the high-level neutralized liquid wastes, salt cake, and sludge are also stored in double walled carbon steel tanks.[2] A leak from an inner tank of one of the storage tanks was contained by the second or backup tank avoiding release to the environs. Improved tanks are also being provided at the SRP.

The Idaho Chemical Processing Plant (ICPP) uses stainless steel tanks to store acidic high-level liquid waste prior to calcining. Calcining produces a less mobile solid waste for interim storage.[3] The tanks at ICPP have maintained their integrity since first put into service in the early 1950's.

Two types of waste tanks are used at the Nuclear Fuels Services (NFS) plant at West Valley, New York.[4] The largest volume of high-level waste at NFS is a neutralized waste contained in a carbon steel tank. A smaller stainless steel tank is being used to store acidic waste.

Although Allied General Nuclear Services (AGNS) plant has not been used to process commercial fuel to date, the storage incorporated into the facility is for acidic high-level liquid wastes in cooled stainless steel tanks.

A survey of European and other nuclear fuel processing countries indicates that all use cooled stainless steel tanks to store acidic liquid waste solutions.[5] Their experience with the integrity of their tanks is similar to the ICPP, i.e., no leakage.

HISTORY OF HIGH-LEVEL WASTE TREATMENT

Methods to solidify high-level nuclear wastes have been under development for about three decades (see Table I). During the 1950's when the nuclear field was emerging from its war time status scientists at the national laboratories and elsewhere were evaluating clay and ceramic products[6,7,8] for permanent storage; this effort, though, was not large. Also methods to solidify the waste

as a calcine, and as a salt cake and sludge were under development.[1,9] By 1963 a full-scale fluidized-bed waste calciner was placed in operation at the Idaho Chemical Processing Plant to demonstrate high-level waste solidification.[10] This plant, still operating today, has calcined more than 3.8 million gallons of waste. During this time Hanford and Savannah River started converting their waste to salt cake. Other solidifcation methods were also being examined during the late fifties and sixties throughout the world including nephaline glass in Canada,[11] borosilicate glass in Britain, France, and the U.S., and metal matrices in the U.S. and Belgium.[12,13] Also during the 1960's pot and spray calcination plus phosphate glass processes were tested at Battelle Northwest Laboratories using radioactive spiked waste.[14]

From about 1969 to 1973 the AEC waste management program in the U.S. underwent major reevaluations which resulted in an expanded federal program. A consolidated program was started at the national laboratories to investigate solidification of specific defense and potential commercial wastes for final repository disposal. Many potential waste forms were examined, such as glass, sintered ceramics, metal matrices, concretes, pellets, and others.[13,15,16,17,18,19] By 1977 the U.S. declared a moratorium on reprocessing spent commercial fuel, and development to solidify commercial waste slowed. Today in the U.S. the major emphasis is on developing processing systems to solidify existing defense waste for final disposal. In France and elsewhere the emphasis is on reprocessing spent power reactor fuel and vitrifying the reprocessed waste, as exemplified by the vitrification plant currently in operation at Marcoule, France.[20]

TABLE I

HISTORICAL HIGHLIGHTS OF HIGH-LEVEL WASTE HANDLING METHODS IN THE UNITED STATES

1944-Present	Acid or neutralized liquid wastes stored in tanks.
1950-1960	Preliminary research on calcine and clay, ceramic glass and metal matrix forms.
1960-1970	Pot and spray calcination and phosphate glass tested with radioactive wastes. Studies on borosilicate glass.

TABLE I

HISTORICAL HIGHLIGHTS OF HIGH-LEVEL WASTE HANDLING METHODS IN THE UNITED STATES
(continued)

1963-present	Plant-scale fluidized-bed calcination of defense waste.
1965-present	Salt cake, sludge storage in tank.
1973-present	Glass and alternative waste forms development on cold and radioactive pilot scale.

CANDIDATE WASTE FORMS

Of the many forms being developed (Table II) on an experimental basis, borosilicate glass is a leading candidate for final disposal of high-level waste. This type of glass has been successfully demonstrated to not only incorporate many types of defense waste, but also potential commercial waste. Extensive data have been collected on its properties,[19] and radioactive production has been demonstrated by at least two methods.[20,21]

A process to incorporate waste forms is the metal matrix.[22] Glass beads or pelleted ceramics are preferred products to cast in lead, aluminum, or other metals. Most advanced on a pilot-plant scale is a molten metal casting process using lead. The metal matrix serves only as a means of encapsulating previously treated waste.

Supercalcine,[18] a product developed to solidify future commercial waste is the most developed of the ceramic waste forms. It is prepared by adding selected chemicals to a liquid waste to form desired crystalline structures on calcining or heat treating. Supercalcine is often pelleted and methods of coating the pellets with pyrolytic carbon and alumina or glass frit are being examined on a pilot scale. Supercalcine has not been extensively tested for defense wastes.

Within the last two years an increased effort has been made to develop crystalline ceramic waste forms for both defense and commercial waste. Often, these efforts are directed towards producing products with crystalline structures similar to those proposed for SYNROC.[23] Laboratory-scale development and characteriza-

tion of SYNROC and related type waste products is in its early stages. No pilot-scale methods have been developed to process this type of product.

Other ceramic related products are also being investigated on a laboratory-scale. These include cermets,[24] a product prepared by using selective additives and controlled heating in a reducing atmosphere to form a highly integrated crystalline oxide-metal matrix. Glass ceramics similar to those produced in industry are also being tested as a waste form.[16]

TABLE II

POTENTIAL HIGH-LEVEL WASTE FORMS

In-Place Solidification Forms	Concrete, rich clay, polymers
Concrete Forms	Normal concrete, polymer concrete, hot pressed concrete
Calcine Forms	Direct calcines, pelletized calcines
Glass Forms	Low-silica glass, high-silica glass, phosphate glass
Ceramic Forms	Supercalcines, SYNROC, mineral ion exchangers, others
Matrix Forms	Metal matrix with glass or ceramic marbles, cermet, multi-barrier and coated particle forms

THERMAL PROPERTY CONSIDERATIONS FOR STORING SOLIDIFIED WASTE

Since heat is generated during the decay of radioactive materials, the disposal temperature and the temperature stability of candidate waste products must be carefully considered. Experimental evidence has shown that most waste forms are not stable at high temperatures ($\sim 350^{o}C$) and pressures. The lack in defining a limiting temperature for repository storage or for the waste form has created a disagreement or misunderstanding as to the long term stability of candidate nuclear waste forms. The temperature of a waste product is dependent on its radioactive concentration, its conductivity, and very important, the conductivity of its surroundings. Radioactive concentration in a waste product can be con-

trolled, for example, by diluting the waste with nonradiocative materials. All solidified waste forms dilute the radioactive concentrations. Second and perhaps most important, radioactivity decays very rapidly during the first few years after reactor fuel discharge. Therefore the decay time before solidification and placement in a repository greatly influences waste product temperature.[25]

By combining proper selection of radioactive concentration and decay time with temperature stability limits of the product and repository (sometimes considered to be 250°C), thermally safe disposal should be practical for many candidate waste forms. A proposed Swedish repository concept utilizes dilution of the waste form, decay time prior to fuel reprocessing, and an interim storage prior to terminal disposal.[26] Radioactive heat versus decay time of a reference high-level commercial waste is shown in Fig. 1. The heat generation rate drops by about a factor of 25 during the first ten years from reactor discharge, and decreases by about another factor of 3 between ten and thirty years. After 30 years, heat generation is cut in half about 40 years, until the Cs and Sr is mostly decayed. Because nuclear waste rapidly decreases in heat output, most products can be placed in a repository at temperatures considered safe for both the product and the repository within reasonable time spans.

CURRENT ACTIVITIES

The abundance of activities in high-level waste management accurately projects the urgency of producing an acceptable system to dispose of high-level nuclear wastes. Every country with operating nuclear power reactors has a major program to develop an overall strategy for waste disposal. The two most prominent types of programs are waste solidification and repository qualifications. In the U. S. high-level waste management program, the defense wastes consume most of the effort. In other countries the major efforts are on programs for handling of high-level wastes from power reactors. The programs are extremely diverse consisting of chemistry, engineering, manufacturing, geology, physics, laboratory, pilot plant, and full-scale plants. It is therefore impossible to cover every laboratory, university, research facility, or government facility. Those presented here only highlight some of the activites.

The French Vitrification Demonstration Plant (AVM) at Marcoule must be considered to be the leader in waste solidifcation

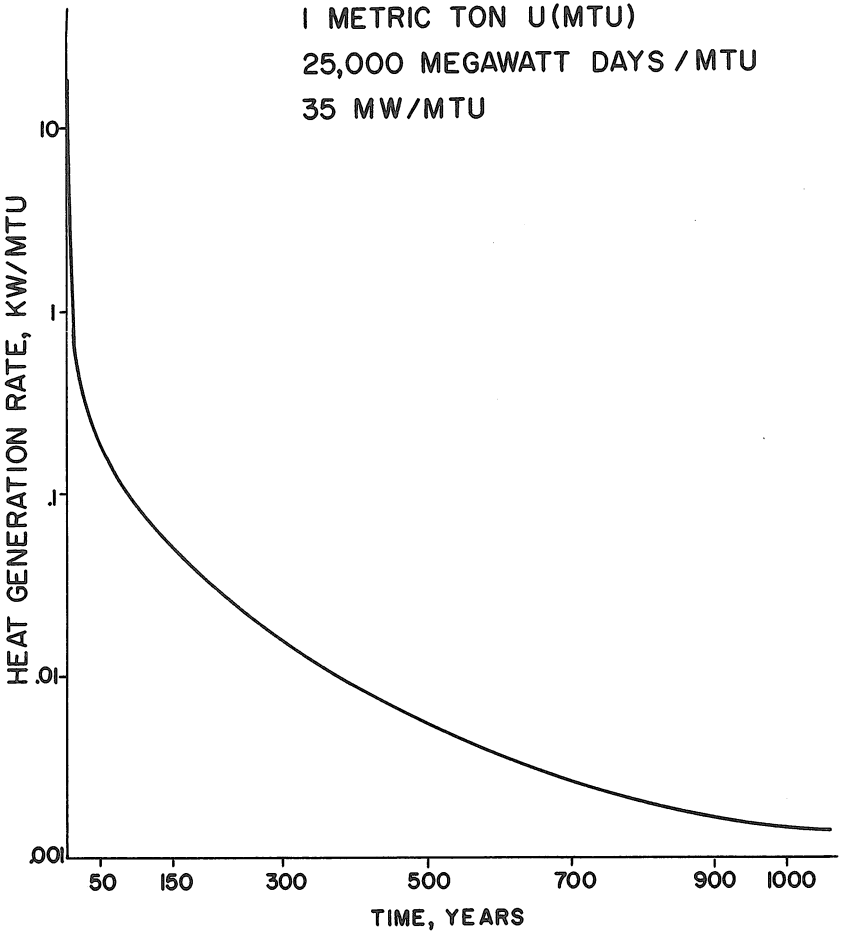

FIGURE 1. HEAT GENERATION RATE WITH NUCLEAR DECAY TIME

demonstrations. The AVM plant has produced at least nine tons of waste glass. The performance characteristics of the large-scale equipment are being evaluated to improve operating reliability.

A recent successful demonstration of a spray calciner coupled to a glass melter was reported earlier at this meeting (see W. F. Bonner, "The Nuclear Waste Vitrification Project Completion, Battelle Pacific Northwest Laboratory").

Fluidized-bed calcination of high-level wastes performed at the Idaho Chemical Processing Plant at the Idaho National Engineering Laboratory is another example of on line waste solidification. This calcined waste (approximately 1750 m^3) has been sampled by coring down through two of the storage bins and has been shown to be mobile and retrievable.

The largest single program in the U.S. for waste solidification is being carried out at the Savannah River Plant. Testing of glass making equipment based on previous work done at other facilities is given a high priority, since it is considered by Savannah River to be the reference process for final fixation of the waste. The major cost, by a large margin, of the proposed Savannah River process is the separation of certain fission products from the salt cake. Extensive pilot scale work is being performed on sludge and salt cake retrieval, ion exchange, evaporation, calcination and vitrification.

A separation and solidification plant is also planned at Hanford. In many respects the wastes at both Hanford and Savannah River are similar, so this work will be mutually beneficial.

Vitrification developments have been spearheaded by the Battelle Pacific Northwest Laboratory. Much of the effort is presently being centered on improving equipment performance and determining waste glass characteristics.[19]

Many universities, particularly those with strong ceramic and geology departments, participate in contract work with DOE in the basic sciences.

The concept of disposal in stable geological formations has also expanded significantly. Originally, emphasis was on disposal in salt formations, but more recently has extended to granite, basalt, tuft, and shale. The Swedish effort to characterize a potential repository is often considered to lead the various programs. It is interesting to note that those countries

which seem to have fewer resources tend to be more objective and productive in this work; the Swedish and the French are good examples.

The work in Sweden at the Stripa Mine on permeability and thermal conductivity has been in cooperation with the United States. Other nuclear power generation countries also have geological evaluation programs. In terms of cost, the U.S. program is the most expensive. Work at Lyons, Kansas, provided some data on the radiation doses; but more recent repository doses; but more recent repository evaluations have been centering at Carlsbad, New Mexico, the basalt project at Hanford, Washington, and the Nevada Test Site. The Federal Republic of Germany, France, Belgium, United Kingdom, USSR, and many other countries are also actively examining the potential of geological repositories. The compilation of K. M. Harmon[5] is an excellent accumulation of worldwide activities.

The use of multiple barriers is being emphasized to provide conservative designs for additional isolation in repositories. This emphasis is becoming wide spread both in Europe and the U.S.

In assessing current activities, the lack of established practical criteria for high-level waste disposal presents the perception of a shotgun approach. The lack of even a demonstration repository in the U.S. is unquestionalbly weakening the acceptance of the nuclear power option. The highly focused program in Sweden lends confidence to the ability of the technical community to provide an acceptable waste management solution. The accomplishments of Swedish Repository studies on permeabiliy and heat transfer, the French success in producing glass billets in a production facility, the Canadian backup data on the exposed waste glass at Chalk River, and the extensive studies in the U.S. on the stability of waste glasses are all positive results and must be integrated into an acceptable disposal system.

POTENTIAL FUEL PROCESSING AND WASTE SOLIDIFICATION SCENARIO

A proposed scenario for fuel processing and waste management is shown in Fig. 2. As a balance between safety and economics, the spent fuel elements would be stored for five years prior to reprocessing to allow short-lived radionuclides to decay. This provides a fuel for processing that is substantially reduced in both radioactivity and heat generation rate. Storage of spent fuel in water ponds has been shown to be safe over long time frames.

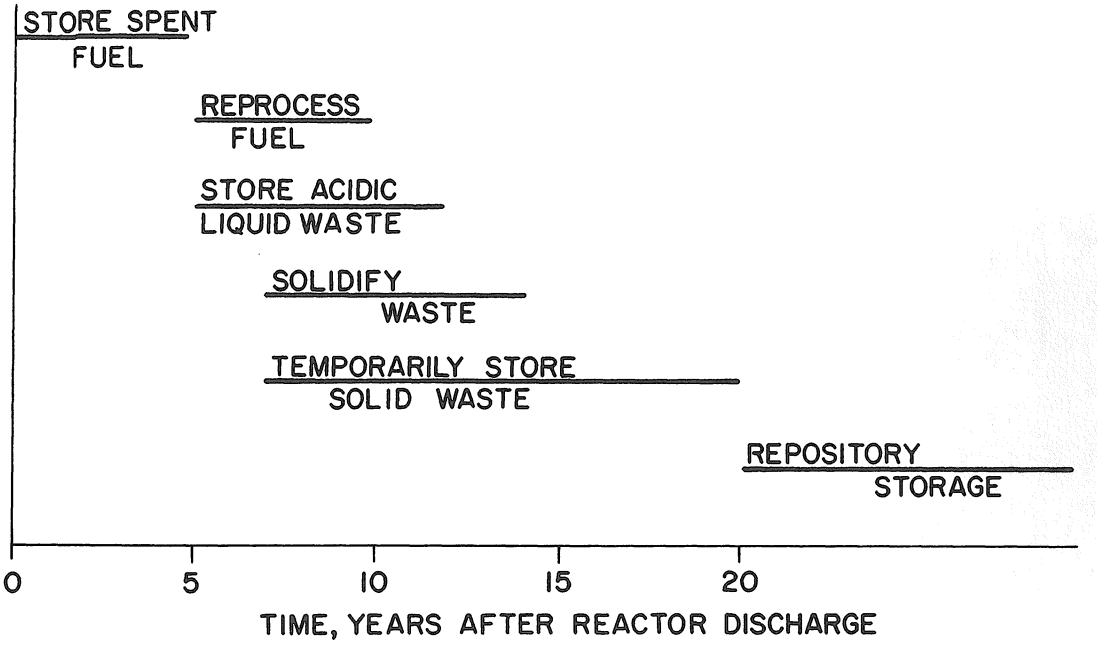

FIGURE 2. PROPOSED SCENARIO TO MANAGE PROCESSED HIGH-LEVEL WASTE

The reprocessing wastes would then be best stored as an acidic liquid in near surface stainless steel tanks for up to two years. The temporary liquid storage would provide reasonable operational storage without large costs as liquid storage is expensive.

The liquid waste would be converted to a final solid waste form, canisterized, and stored temporarily in engineered facilities for 10 to 20 years prior to placement in a final waste repository. By this means the waste is converted to a solid within reasonable time periods and storage temperatures in a repository are minimized.

This scenario is based on proven and safe concepts and could be put into practice today. Interim solid storage allows the time required to qualify a final repository.

Basically all the U.S. high-level wastes from both defense and potential commercial fuel reprocessing by virtue of cooling time and dilution can be made into low heat generation waste forms. The cooling time of power reactor spent fuels will be at least 10 years in the forseeable future before they are reprocessed. The waste from these fuels, therefore, will also be relatively low in heat generation. Until some time in the future when commercial fuel reprocessing catches up with spent fuel production, heat generation by solidified wastes will not approach the limits of potential repository storage temperatures.

CONCLUSIONS

Storage of power reactor spent fuel is necessary at present because of the lack of reprocessing operations, particularly in the U.S. By considering the above solidification and storage scenario, there is more than reasonable assurance that acceptable, stable, low heat generation rate, solidified waste can be produced, and safely disposed.

The public perception of no waste disposal solutions is being exploited by detractors of nuclear power application. The inability to even point to one overall system demonstration lends credibility to the negative assertions. By delaying the gathering of on-line information to qualify repository sites, and to implement a demonstration, the actions of the nuclear power detractors are self serving in that they can continue to point out there is no demonstration of satisfactory high-level waste disposal.

By maintaining the liquid and solidified high-level waste in

secure above ground storage until acceptable decay heat generation rates are achieved, by producing a compatible, high integrity, solid waste form, by providing a second or even third barrier as a compound container and by inserting the enclosed waste form in a qualified repository with spacing to assure moderately low temperature disposal conditions, there appears to be no technical reason for not progressing further with the disposal of high-level wastes and needed implementation of the complete nuclear power fuel cycle.

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