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DEVELOPMENT OF SOLID RADIONUCLIDE WASTE FORMS
IN THE UNITED STATES

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INTRODUCTION

Until fairly recently, there were really two nuclear waste forms:

- Solid waste, which was by definition what went into a burial ground even if the waste happened to be mostly liquids, and
- Liquid waste, which was by definition what went into a waste tank even if the waste happened to be mostly solids.

In detail, of course, the solid wastes subdivided into beta-gamma wastes, transuranic wastes, medical wastes, reactor wastes, R & D wastes, and what have you, while the liquid wastes subdivided into high-level wastes, intermediate-level wastes, acid wastes, alkaline wastes, etc. However, the overall division remained clear.

Obviously this situation no longer holds. Gaseous wastes have been added to the list, and a new nuclear waste category (spent fuel) has been invented which doesn't fit into any of the previous categories. More important, the implied end-points for waste disposal have changed. Except in circumscribed cases involving high localized, very low-level, or very short-lived activity, simple burial or simple tank containment of the wastes is no longer considered adequate. Instead, the end-points

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now being looked at include geologic, ocean bed, and even ice sheet or space repositories.¹ Transmutation of the waste radionuclides is another further-out possibility.² All of these new end-points require extensive reworking of the wastes - to reduce their volume, to prepare them for shipment from the generating point to the disposal point, and to provide additional barriers against radionuclide dispersion. This extensive reworking of the wastes requires a new classification in terms of the final waste forms. The candidate forms are surveyed in this paper.

I will first address the types of waste forms, then to who is working on them, next to the waste form requirements, and finally to waste form selection.

WASTE FORM DESCRIPTIONS

Encapsulated Waste Forms

For many of the solid wastes, the radionuclide levels are so low or they are so tightly fixed to the waste that these wastes can be directly packaged with, at most, a volume reduction and surface decontamination.³ The same approach may also apply to spent reactor fuels, which were, of course, originally fabricated in very high-integrity forms to withstand the extremely rigorous reactor conditions.⁴ The next stage beyond direct packaging is encapsulation of the waste in a suitable binder material. The binder serves both to impede leaching and to prevent mechanical dispersion of small waste particles. It may also provide a contamination-free surface. Technology for such encapsulation is well advanced in the low-level and transuranic waste programs. It is in general limited to these programs. The two binders in common use are concrete and organic polymers. Their advantages and disadvantages are discussed below.

Concrete Binders.⁵ The concretes are readily formed with existing technology and present few processing problems except incrustation of the mixing equipment. For the most part, concrete binders offer excellent fire resistance, good adhesion and shielding, and intermediate leach rates and radiation resistance. Their main disadvantages are weight and the potential for high gas release, both from irradiation and hot fires.

Organic Binders. The organic binders include the natural bitumens,^{6,8} urea formaldehydes,^{7,8} thermosetting polyester-styrene resins, and vinyl esters.⁸ They are easy to use, lightweight, and have excellent leach resistance. However, their radiation resistance is limited to 10^{10} rad or less, and they may be flammable or chemically unstable particularly if oxidizing agents are present. There are also questions on the very long-term stability of any organics.

Although current Department of Energy (DOE) planning is aimed at placing essentially all high-level and transuranic wastes in geologic repositories, it may turn out that, for some of the older defense wastes, the risks of removal and treatment are greater than those of immobilizing the wastes in place. The same considerations apply even more strongly for low-level and intermediate-level wastes. Several options including concrete, clay additives, and calcines have been considered for inplace solidification of liquid wastes under these conditions. These options are described below.

Concrete. The Oak Ridge National Laboratory has for a number of years been conducting a successful program to dispose of its intermediate-level wastes by grouting them into special concrete formulations and hydrofracturing the grout into shale formations below the laboratory.⁹ Although limited to particular geologies, this is an inexpensive and straightforward disposal method that has given uniformly good results to date. Concretes have also been given limited consideration for inplace solidification of the high-level wastes in the Hanford tanks as part of a tank "hardening" program.

Clay Solidification. Clay additives have also received limited attention as a solidifying agent for the Hanford high-level wastes, offering the possibility of lower waste volumes and reduced reaction temperatures than with concrete solidification.¹⁰

Calcines. Calcines are formed by firing the wastes to a mixture of (mainly) metal oxides at 300° to 700°C.¹¹ Calcines can be either an intermediate in the preparation of more advanced forms or a waste form in their own right. They have the disadvantage of water solubility and of small particle size, which can lead to high dispersability. However, calculations at the Idaho Chemical Processing Plant, which has pursued a long-term program of calcining its wastes, indicate that the risks of storing the calcine under their geologic conditions are small and can be further reduced by pelletizing and chemically treating the calcine. A limited program is investigating these possibilities.

Glass and Ceramic Waste Forms

Except for the very limited effort on inplace solidification of older defense wastes described in the last section, essentially all of the effort on high-level waste forms both in the U.S. and abroad is aimed at chemically binding the waste atoms into a high-integrity lattice which will strongly resist mechanical dispersion, volatilization, or leaching. The same approach is

also expected to be applicable to other "chemical" waste such as incinerator ash from burning flammable low-level and transuranic wastes, decontaminant solutions, and treated cladding hulls. The simplest and most studied high-integrity waste lattices are the ceramics and glasses. Borosilicate glass in particular has received extensive development, is in commercial use for waste management by the French, and is the reference material in most other national programs for high-level waste forms. The various glasses are described below, and the ceramics currently being investigated for the waste forms are discussed immediately after.

Phosphate Glass.¹² Phosphate glass has received considerable attention as a waste form. It is produced by mixing the waste with phosphorus pentoxide or phosphoric acid and melting the mixture at about 1100°C. Phosphate glasses will incorporate about the same waste loadings as borosilicate glasses and are better at accommodating molybdates and sulfates. Waste leachability and volatility from the phosphate glasses are also similar to those for borosilicate glasses. However, two major deficiencies have largely ended work with the phosphate glasses. First, they are extremely corrosive, so that almost the only materials that can be in contact with the melt are platinum and some refractories. Second, the phosphate glasses devitrify at about 400 to 500°C to produce a 1000-fold increase in leachability.

Borosilicate Glass.¹³ Borosilicate glass is currently the best developed waste form for high-integrity radionuclide containment. Typically a borosilicate waste glass might consist of about 20 to 30% waste oxides, 30 to 40% silica, 5 to 10% boron oxide and 10 to 15% alkali oxides, plus other additives. Formation temperatures are in the range 1100 to 1200°C for these compositions, and two main fabrication methods are in use: external melters from which the waste glass is poured into its final form, and in-can melters in which the waste glass is formed in place in the final canisters. Large glass monoliths up to three-feet in diameter and twelve-feet high can be formed with either external or in-can melters. The external melters can also produce marbles for use in matrix forms. The advantages of borosilicate glass waste forms are low leachability, excellent radiation stability, low volatility, relative insensitivity to composition, and, in the canisters, good mechanical strength. The primary concerns about borosilicate glass are leaching and chemical attack by high-temperature, high-pressure water or brine, devitrification, and cracking. These concerns are quite serious for high-heat, high-level waste loadings for which surface temperature in the repositories may reach 250 to 350°C. Such temperatures will definitely lead to short lifetimes for the glass in contact with pressurized repository water. However, the low-heat, high-level wastes from the U.S. defense program can easily be stored at surface temperatures of no more than

100°C. At these temperatures, borosilicate glass has demonstrated excellent characteristics in all tests to date. Similarly, as is being done in the French program¹⁴ for incorporating their reprocessing wastes in borosilicate glass, it is possible to decay high-heat wastes for 40 to 50 years before final emplacement and again to achieve high predicted repository lifetimes with 100°C surface temperature.

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High-Silica Glass.¹⁵ High-silica natural glasses (obsidians and tektites) are known to have persisted for long periods in both terrestrial and lunar environments. However, these glasses are formed at about 1600°C, a temperature high enough to drive off most of the ruthenium and cesium radionuclides from the wastes. Thus, the key to producing these glasses as waste forms is to find low temperature processes for producing them. Several proprietary processes are being investigated for this purpose, but the products have not yet been thoroughly characterized.

Glass-Ceramics.¹⁶ One form of glass ceramic can be made by sintering or hot-pressing the mixtures of waste and glass frit at comparatively low temperatures rather than melting it as in normal glass-making practice. The advantages are reduced radionuclide volatilization and chemical corrosion, although at some loss in form integrity. The more common forms of glass-ceramics are formed by adding nucleating agents to the glass melt and then cooling the glass in a controlled manner so as to produce an assembly of small crystals held together by a glassy phase. The chief advantages of the glass-ceramics are good thermal shock resistance, low leach rates, and high temperature stability. Their disadvantages are complex processing and relatively small sizes.

Hot-Pressed¹⁷ and Special Concretes.¹⁸ Studies at the Savannah River Laboratory showed that normal concretes give an inferior product at higher cost than borosilicate glass for immobilizing high-level wastes. Accordingly, all current studies of concrete high-level waste forms are aimed at advanced forms. Two possibilities are being explored. The one is to hot-press the concrete so as to eliminate voids and non-chemically combined water. The aims are to decrease the waste form volume, decrease the effective leaching areas, and reduce steam volatilization in hot fires. The other approach is to preform the waste into particularly leach-resistant particles (such as "Supercalcine") before incorporating it into concrete. The aim is for these particles to retain enough chemical integrity to improve the concrete leach rates.

Clay Ceramics.¹⁹ Adding aluminum silicate clays such as kaolin or bentonite to the waste produces an insoluble cancrinite-type material. This material can be fired to a nepheline-type material. Typically, the waste is mixed with the clay and water, extruded into strings, chopped into pellets, dried, and fired at

about 1200°C. The product is markedly inferior to glass or the artificial minerals as regards leaching, dusting, surface area, etc. Accordingly, little attention is being given these forms.

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Mineral Waste Forms

Although limited archeological data are available on some glasses and ceramics for periods of up to several thousand years, most of the materials data for long time-periods are in the geologic record. Accordingly, there have been several efforts to model waste forms after geologic minerals, particularly after those minerals which have experienced natural radioactive decay without extensive lattice damage. The two major artificial mineral systems which have been examined in this regard are the silicates as exemplified by the Supercalcines and the titanates as exemplified by SYNROC.

Supercalcine.²⁰ Pennsylvania State University working in cooperation with Pacific Northwest Laboratories has developed a series of waste forms they call Supercalcines. The Supercalcines are formed by calcining the waste with suitable additives such as Ca, Al, Sr, and Si oxides and then heating the calcines at 900 to 1250°C to produce a polycrystalline form. Typical crystal phases include pollucites, monazites, apatites, scheelites, sodalites, fluorites, spinels, and corundums. The Supercalcines offer the advantages of very high waste loadings, up to 60%, good leach resistance, high thermal stability, and if properly formed, high thermodynamic stability. They can be directly produced as small particles suitable for the matrix forms discussed in the next section or they can be pelletized or hot-pressed to larger forms. Three main questions arise for the Supercalcine waste forms. The first concerns the multiplicity of crystalline species and the difficulty of assuring that an equilibrium assemblage of the desired species has been formed. The second question concerns the stability of the various crystals under heat, radiation, transmutation, and leaching stress. Finally, the third question concerns the stability of pollucite crystals used to immobilize ¹³⁷Cs. Pollucite has been shown to have high leach rates in hot brines and in water at high temperatures and pressures.

SYNROC.²¹ Prof. Ringwood at the Australian National University has developed several assemblages of synthetic titanate minerals, which he calls SYNROC. The current reference SYNROC composition is a mixture of fairly large (several millimeter) crystals of hollandite, perovskite, and zirconate. Typically, the SYNROCS are formed at very low waste loadings, 5 to 10%, but they may be able to accommodate quite high loadings of the alkaline defense high-level wastes by using the inert alumina and ion phases in the waste as carriers for the SYNROC particles. SYNROC can be formed by melting (1600°C), hot isostatic pressing

(1100 to 1350°C), and possibly by hot sintering at about 1000 to 1200°C. SYNROC has an outstanding resistance to high-temperature leaching, showing no changes at 800°C and 1000 bar for 24 hours. It also has a high density and can be expected to show high thermal and radiation stability. Relatively little work has yet been done on SYNROC, and a large number of questions remain about its ability to handle a large variety of waste species, about its long-term thermal and radiation stability, and about its fabrication techniques.

Titanates, Niobates, Zirconates.²² Sandia Laboratories have developed a series of hydrous-oxide inorganic ion exchangers with the proposal that they be used to concentrate waste radionuclides and then be sintered into low-leachability ceramic waste forms. Mixing the radionuclide-loaded resins with about 30% glass and sintering at 900 to 1100°C at pressures of 7 to 14 MPa gives low leachability products. These products have not been well characterized, and a large number of questions remain about non-equilibrium phases. Also the ¹³⁷Cs is in a pollucite phase, which may be leachable under hydrothermal repository conditions.

Matrix Waste Forms

Many of the waste forms may be difficult to fabricate in large monoliths, and, even for those where monoliths are possible, quality assurance testing of the product will in general be easier for one- or two-inch pieces than for very large pieces. It may, therefore, be necessary to go to matrix waste forms simply for mechanical reasons. Other more positive reasons are that the matrix material can offer another layer of protection and, if it is a metal, can offer better heat transfer. Multi-barrier materials in which the waste particle is coated with additional protective layers before incorporation in the matrix are still further elaborations. For all the matrix forms, the trade-off is between the advantages of the matrix configuration against the greater waste surface areas and processing complexities involved in going to the matrix configuration. The advantages and disadvantages of the matrix forms are discussed below.

Metal Matrices.²³ Most of the waste forms discussed to date such as calcines, concretes, glasses, ceramics, and artificial minerals can be dispersed in a metal matrix for better heat transfer, reduced frangibility, easier sampling, and additional leaching barriers. Low-melting alloys such as Pb-Sb/Sn or Al-Si can be cast around the waste particles, while higher melting metals can be sintered around the particles at temperatures of about two-thirds their melting temperature. Aside from the process complexity, the main questions for these forms are securing void-free matrices, loss of low-melting matrices in hot fires, radionuclide volatility with high-melting matrices, and the

corrosion lifetimes of the matrix metals. The matrix forms will also, by definition, have lower waste loadings than the direct waste form.

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Multibarrier Forms.²³ Perhaps the most complex waste forms are made by coating the waste particles with impervious materials such as carbon, alumina, or silicon carbide before placing the waste particles in a matrix. Such coatings provide additional barriers against waste leaching and also allow the use of high-temperature matrix formation processing by reducing radionuclide volatilization. Concrete, sintered ceramics, glasses, and other materials in addition to metals can be used as the matrices when coated particles are used. The main questions about the multibarrier forms are their high complexity, both as regards costs and as regards producing an acceptable product. The product quality questions are particularly acute, because most of the existing particle coating technology involves coating very small particles having very large surface areas. A coating failure of such a product could greatly increase radionuclide release over that for a monolith.

Cermets.²⁴ Cermet high-level waste forms are a particular metal matrix form in which very fine waste particles are dispersed in the metal either by in situ reduction of the metal from the waste, or by in situ precipitation of the waste from the metal. Ideally, these forms should be easier to form than the other matrix forms and should have very good bonding between the metal and waste. The main questions on this form versus the other metal matrices are the small size with resultant large surface area of the waste particles, and the fact that their composition may not be as closely tailored as if they had been formed in a separate step.

Cladding Waste Matrices. Most of the current proposals for reprocessing fuels from commercial light water power reactors involve shearing the fuel assemblies and leaching out the uranium oxide, leaving the Zircaloy cladding hulls and other hardware as waste. The hulls can be treated to recover the radionuclides for incorporation in the waste forms discussed to date, or the hulls themselves can be used as the waste particles to be incorporated in a suitable matrix. Relatively little work has been done on the cladding waste matrices; but some of the matrix materials being considered include lead alloys, sand, concrete, bitumen, and glass (possibly even the waste glass material).^{8,25} Stability and containment problems still need to be examined for all these materials.

Gaseous Waste Forms

A number of waste radionuclides including ^{129}I , tritium, and ^{14}C (and even ^{106}Ru and ^{137}Cs in high temperature processes) potentially can exist in gaseous form. However, it is believed

that most of these materials can either be fixed in solid form for incorporation in the waste forms already discussed, or, in the case of the shorter-lived radionuclides such as tritium and ^{106}Ru , simply held for decay. The noble gas ^{85}Kr , which is presently being released to the atmosphere without exceeding discharge limits, may, however, have to be specially stored for a hundred years or so if a full-scale nuclear fuel reprocessing industry develops. Possible waste forms for ^{85}Kr , still being developed, include pressurized gas cylinders, glass, concrete, metal or plastic "foams", molecular sieves, and clathrates.

Canisters and Engineered Barriers

The waste forms are normally housed in a canister which provides a contamination-free handling surface, adds mechanical strength, and supplies extra containment during shipping, handling, and storage. This canister may in turn be surrounded by additional isolation materials, secondary canisters, and other engineered barriers. In some cases, the canisters and engineered barriers can be designed to provide radionuclide containment equivalent to that of the waste form itself. They are also, in general, much less expensive to prepare than the waste forms because most of the work, except final installation, can be done in a nonradioactive environment. Hence, it is important to consider these barriers as an integral part of the waste form development.

The engineered barriers, some of which may be installed as part of the repository, also form the buffer between the waste form and the repository. Here they act to restrict and condition any flow of water from the repository to the waste form, and similarly to restrict and condition any flow of leached radionuclides from the waste form to the repository.

WASTE FORM DEVELOPERS IN THE UNITED STATES

Currently, the largest expenditures for waste form development both in the United States and in the rest of the world are being made to engineer borosilicate glass as a possible waste form for high-level waste and to engineer slagging pyrolysis incinerators for transuranic waste. However, the U.S. is also spending about \$15 million a year to develop and characterize other waste forms which may offer improved radionuclide immobilization. The immediate target for this effort is to select, by 1984, the waste form for the initial DOE high-level waste immobilization plant, the Defense Waste Processing Facility (DWPF), at the Savannah River plantsite. Studies are also expected to continue over a much longer period to take advantage of developing technology for second and later generation waste forms.

The U.S. programs for waste-form development are widely disposed through the waste processing sites, the DOE national laboratories, industrial laboratories, and universities in order to secure the widest possible input. Table 1 lists the major participants in the U.S. high-level waste program.²⁶

Table 1. U.S. High-Level Waste Form Developers

Waste Form	Developer
Concrete (Hot Pressed)	ORNL, PSU
Sintered and Pelletized Calcine	ICPP
Borosilicate Glass	PNL, SRL
Supercalcine	Rockwell, PSU
SYNROC	LLL
Titanate Ion Exchangers	Sandia
Supercalcine Coating and Matrices	PNL
Glass-Marble/Metal Matrices	ANL, PNL
Cermets	ORNL

These U.S. development programs are the responsibility of the Division of Waste Products in the DOE Office of Nuclear Waste Management. The Division of Waste Products has in turn established lead offices for this work at the Savannah River Plant for high-level waste, at Rocky Flats for transuranic waste, and at EG & G and ORNL for low-level waste. Close coordination is also required with the transportation lead office at Sandia and the Office of Nuclear Waste Isolation at Battelle-Columbus.

WASTE FORM REQUIREMENTS

The waste forms must be considered as one part of a total nuclear waste disposal system. Their primary role within this system is to provide the initial barrier against radionuclide release. Here the overwhelming emphasis has been on reducing the availability of the waste radionuclides to water leaching when the wastes are disposed of in a repository. Current multi-barrier philosophy is, in fact, that the waste form should provide a completely redundant backup to the repository barriers until the waste hazard has been greatly reduced by radioactive decay. The Nuclear Regulatory Commission has informally proposed a goal repository integrity period of 1000 years for the waste form plus its engineered barriers;²⁶ 1000 years is roughly equivalent (for reprocessed waste) to decay to the radioactivity level of the ore from which the reprocessed fuel was originally

prepared. Other possible goals are 300 to 500 years to allow for ^{137}Cs and ^{90}Sr decay, or 10,000 years or more, as suggested by EPA, to allow for partial decay of the very long-lived nuclides.

Overall requirements on the waste form include low dispersability and good impact resistance in transport, storage, and sabotage situations; low radionuclide volatility in hot fires; long-term stability; low sensitivity to waste variations; good waste density loadings; and technology availability. Process factors are equally important and include processing safety both to the operator and the public, the ability to produce a fully licensable product at good yield, acceptable costs, and minimum secondary waste generation.

WASTE FORM SELECTION

Plans for waste form selection in the U.S. are discussed below.

Waste Form Characterization

At the present stage of the U.S. waste forms programs, the main emphasis is on defining and characterizing the forms. This will enable reliable comparisons among the different forms, and against the requirements being established by the process, transportation, and repository groups on the one hand and by the regulatory agencies on the other. It is expected that most of the characterization will be performed by the forms developers themselves; however, in order to systemize the process, the Battelle Pacific Northwest Laboratories have been designated to prepare standard procedures, perform quality assurance testing, and publish certified waste form data for the U.S.

Waste Form Production

The worst danger of a paper such as this one is to lose sight of the fact that waste-form development is not an end in itself. Its purpose is to dispose of the nuclear wastes at the earliest reasonable time schedule. Therefore, once the characterization work has supplied enough data (the goal date in the U.S. is 1982), the next phase is to enter into detailed engineering development of a few selected waste forms. After about two more years, the data should be available to select (by a suitable technical regulatory, legislative, and public process) the final waste forms for the actual waste disposal plants. As mentioned earlier, the target data for final selection of the waste form

for the initial U.S., high-level, waste-form plant at the Savannah River plantsite is in 1984. France has already made its decision for borosilicate glass, and a number of other countries seem close to a decision. Hopefully, then, we can see the waste forms work culminate within about 10 years.

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