# SURFACE ENCAPSULATION PROCESS
# FOR MANAGING LOW-LEVEL RADIOACTIVE WASTES

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## ABSTRACT

Current processes for low-level radioactive waste (LLRW) stabilization involve mixing contaminants with a fixative such as cement, asphalt, polyethylene, or vinyl monomers, and subsequently curing the mixtures in containers. These methods give rise to processing difficulties and yield products lacking performance to assure long-term LLRW immobilization. Mixing of LLRW into fixatives is impeded by viscous media and the curing reaction is inhibited by LLRW constituents. Product performance is affected by corrosion of the containers which ultimately expose the cured mixtures to environmental stresses. Our process, termed the "Surface Encapsulation Process," circumvents these problems. A thermosetting fixative is employed that mixes readily with LLRW and is highly insensitive to inhibition in curing. The agglomerated mixtures are further stabilized by encapsulation with seamless jackets of corrosion resistant plastic, such as polyethylene.

In laboratory-scale investigations, feasibility of our technique was demonstrated for managing a broad spectrum of LLRW simulants including ion-exchange resins, beads, and glasses, and sodium salts. Products tested to date meet all relevant NRC and DOT regulations governing waste fixation. The high waste loadings of the products, use of commodity resins, and processing simplicity indicated our process would provide high performance LLRW stabilization at costs that are competitive to those for processes employing state-of-the-art fixatives. An economic analysis based on managing LLRW generated by commercial power plants (-1,000 MeW) substantiates the competitive process costs advantages.

To carry out full-scale management of LLRW by our process, we have completed construction of a novel process demonstration unit under sponsorship of the Department of Energy. The apparatus stablizes LLRW in cylindrical 50-gallon modules by agglomerating waste mixtures and powder molding 1/4-inch thick, polyethylene seamless jackets onto the surfaces of the agglomerate. The apparatus is mobile and has capacity to process 3,000-tons of LLRW per year. It features both agglomerating and jacketing molds mounted on a single frame and process controls that are remotely operable.

## INTRODUCTION

Low-level radioactive waste (LLRW) management favors waste concentration as the means to realize subsequent cost and safety benefits in waste stabilization, transportation and final disposal. The problem is fabrication of high performance stabilization products with residues that stem from LLRW treatment processes. It is difficult to secure the benefits of waste concentration because fabrication cannot employ more than minimal amounts of fixative.

Current LLRW stabilization processes, in our assessment, would not yield high performance products with concentrated wastes. Even at appreciable amounts of fixative, product mechanical performance and resistance to leachate decreased significantly as contaminant loadings were further increased; in these cases cement, silicate, and asphalt were used as fixatives.[1,2,3] The use of synthetic resins was characterized by difficult processing operations due to their high melt viscosities, or to their reaction chemistry that can cause premature gellation and excessive exotherm. Furthermore, where stabilization entails chemical reaction, chemical pretreatment of the LLRW may be required in order to prevent reaction interference. This operation would increase process costs and the amount of fixative used for stabilization.

The Surface Encapsulation Process provides a viable option in waste stabilization. It stabilizes concentrated LLRW cost effectively with amounts of fixative, and high performance products fabricate readily and reproducibly. In our work industrial contaminants and LLRW simulants were stabilized on a laboratory-scale. We employed a fixative that is unique in waste stabilization and novel surface treatment to reinforce the product. Stabilization was found to occur in the presence of a wide variety of contaminants and a broad range of pH. The products were mechanically tough and resistant to harsh leachates. The process was readily applicable to management of LLRW soluble salts and ion-exchange resins; these wastes being considered to be amongst the most difficult to manage. As a result, we designed and constructed an apparatus to fabricate products simulating our laboratory-scale ones in the commercially viable size of 50-gallons.

This paper reviews our process features, product performance and recent work for managing nuclear grades of ion-exchange resins and soluble salts in concentrations greater than that previously reported in the literature. The paper discusses the fixatives and surface treatment materials employed, the process products, performance of laboratory-sized modules, full-scale processing criteria and economics and the apparatus we have recently fabricated to carry out full-scale LLRW processing. The information is arranged under the headings of: 1) Fixative, 2) Surface Treatment, 3) Product, 4) Process, 5) Product Performance, 6) Apparatus, and 7) Cost Benefits.

## Fixative

Our investigative work showed 1,2-polybutadiene and its mixture with powdered polyethylene to have advanced utility in contaminant fixation.[4] These resins thermoset in the curing reaction to yield a chemical structure analogous to cross-linked polyethylene. Such material has excellent chemical stability and withstands mechanical overburden without creep. Unlike synthetic fixatives currently used, the curing reaction of 1,2-polybutadiene proceeds in the presence of a wide variety of contaminants. These benefits are gained due to the resin's hydrophobicity and high functionality. Since the resin will not absorb polar and mineral impurities, these materials do not interfere at the fixation reaction site. The high functionality of the resin gives rise to three dimensional resin networks thereby yielding dimensionally stable products. Thus, chemical pretreatment of the waste is not necessary to structure them for mixture curing.

In contrast to fixatives in which the curing reaction is initiated upon addition of a promoter into the mixture, the curing reaction of a 1,2-polybutadiene is thermally initiated. Thus, currently employed resins cure en toto and generate appreciable exotherms in commercial-size modules. In contrast, mixtures of polybutadiene and LLRW cure incrementally with the surrounding material providing a heat sink. Thus, the temperature of the mixtures in Surface Encapsulation do not exceed their reaction initiation temperature.

Thermally initiated curing and stability at temperatures below threshold also affords other processing advantages. It permits LLRW/fixative blending operations to be carried out without time sensitive constraints such as premature curing of the mixture. It allows the option of storing and inspecting the mixture prior to the formation of stabilized products.

## Surface Treatment

Reinforcement of the fixative/polybutadiene agglomerate is accomplished by fusing 1/4 inch thick polyethylene onto the surfaces. This surface treatment is unique in waste management and offers several performance benefits compared to both state-of-the-art waste management surface treatments. The surface treatment that was developed is lamination with impregnation and it is compared to other surface treatments used in waste management: 1) material impregnation, 2) surface coating, and 3) lamination. Material *impregnation* is exemplified by work carried out at Brookhaven where LLRW are stored in concrete containers. In order to reduce water permeation into the containers, solutions of styrene monomer are applied onto the container and polymerized in situ. Although permeation is reduced, the resultant polymer impregnated cement remains brittle. Examples of surface coatings that are used in waste management include epoxy coated steel drums. Although chemical and corrosion resistance are achieved, no mechanical benefits accrue. Simple lamination is employed in fabricating fiberglass holding tanks. However, the adhesive bonds between layers of substrate are prone to catastrophic failures. Lamination and impregnation of the substrate is unique with respect to the above techniques. It provides a seamless layer of plastic that penetrates into the substrate and causes a 1/4 inch layer of resin above the substrate. The layer acts to reinforce the substrate both mechanically and chemically and also serves as an impervious barrier isolating the substrate from environmental stresses.

Compared to containers used to reinforce state-of-the-art fixative/contaminant mixtures, the Surface Encapsulation jacketing provides enhanced benefits. Containers fail due to corrosion, ineffective closures, and by distortion caused by overburden stresses. The Surface Encapsulation Product is corrosion-resistant and is seamlessly jacketed; i.e., there are no closures. Due to the penetration of the jacket into the waste containing substrate, the waste consignments in the Surface Encapsulation products are load bearing. By using a flexible jacketing resin, the hazardous consignments remain secure even in severely crushed products. The jackets can be readily repaired if damaged.

## Product

The stabilized waste products are characterized by 1/4 inch thick polyethylene seamless jackets fusion bonded onto agglomerates of LLRW concentrated in a matrix of coreacted polyethylene and polybutadiene. The resin jackets provide radiation stable reinforcement of the fixed contaminants, thus yielding waste forms that assure high performance contaminant stability under harsh environmental stresses.

Commercial-size modules that are currently under development are cylindrical and approximately 50-gallon in capacity. The module diameter is approximately 24 inches, similar in diameter to a 55 gallon drum. Thus, the modules can be managed by conventional drum handling equipments. Cylindrical modules were specified also to dissipate heat from modules containing LLRW. Modules can be readily fabricated in other sizes and configurations, such as cubic in order to conserve landfill space. The 50-gallon modules that we are developing will weigh between 300-600 lbs depending on the density of LLRW. Approximately 50 lbs of resin are needed to fabricate the modules; 25 lbs of fixative and 25 lbs of jacketing resin. The fixative and jacket add less than 5% to the waste volume.

## Process

Full-scale processing of LLRW is based on procedures developed in laboratory-size module fabrication and entails three major steps: 1) mixing waste with PB binder, 2) agglomerating the mixture to yield a core, and 3) encapsulating the core with a resin jacket. These processing steps are discussed below.

Mixing wastes and the resin has been carried out in a low-compression laboratory extruder and in a stirred vessel with solvent diluted resin. Mixing undiluted liquid resin obviates the need for solvent recovery in full-scale processing. Because resin melting is not required in mixing, low compression extruders can be utilized which reduces power requirements, screw wear, and is simple to control compared to other extrusion operations used in waste management. In our work, the LLRW and the PB binder are fed to separate input ports at measured rates. In laboratory work, the resin is gravity fed through a metered orifice and the LLRW is transferred by a vibratory feeder. The resin and the extruder barrel are heated to approximately 150°F to reduce resin viscosity and facilitate mixing. Full-scale processing can be accomplished at temperatures as high as 350°F to remove free water in the waste input and achieve maximum volume reduction. Full-scale processes can also utilize batch mixing equipment such as ribbon blenders and mills for waste/binder mixing.

The waste/binder mixture is then transferred to the apparatus where it is first agglomerated and then encapsulated. Agglomeration is carried out by compression molding in which the mold is heated and minimal pressure is applied to the mixture. The shape and size of the agglomerating mold determines the dimensional nature of the final product. As explained earlier, we are currently designing 50-gallon cylindrical modules; however, rectangular modules can be fabricated in order to conserve landfill volume. Molds can be heated directly by electrical heaters or steam or indirectly in curing ovens. Cure time is dependent on thermal conductivity of the waste material. We estimate a cure time of 1-6 hours for commercial-size agglomerates.

In the final step, the waste agglomerate is jacketed with polyethylene by powder molding. The waste agglomerate is loaded into a matched mold, i.e., a mold geometrically similar but slightly larger than the agglomerate. Powdered polyethylene is then loaded into the interstitial space between the agglomerate and the jacket mold and on the top surface of the agglomerate. The jacket is consolidated and bound to the agglomerate by heating the powder so that it melts and fuses and then cooling the melt so that it solidifies. The partially jacketed agglomerate is inverted and additional polyethylene is added over the unjacketed surface and the jacketing is completed.

## Product Performance

Laboratory-size products were evaluated for both mechanical performance and resistance to leachate stresses. Under EPA sponsorship, over 100 laboratory-scale modules with 90% by weight hazardous content were prepared and evaluated. The modules held residues of the following dewatered sludges: electroplating, nickel-cadmium, $SO_x$ scrubber, paint pigments, chlorine production brine, and calcium fluoride.

The following mechanical performance was observed: under uniaxial mechanical compression, modules withstood appreciable pressure before deformation because the consolidated contents distributed the applied load and the matrix resin resisted creep due to its chemically cross-linked nature. Under extreme pressure the modules yielded, but even when severely distorted there was no content loss because their jackets provided flexibility and toughness which functioned to envelope the contents and keep them intact. Under impact by dropping, modules remained intact upon landing on a steel plate. We estimated it would require extraordinary impact to rupture a module, and even then there would be no spillage of waste contents because they are fixed in a resin matrix. In puncturing by pointed steel probe, appreciable force was required to penetrate the jackets because they were both tough and reinforced by being bonded to the surfaces of the resin matrix agglomerated contaminants.

Further evaluations of modules were carried out with respect to their physical and chemical performance. At the Corps, modules were subject to wet-dry and freeze-thaw conditions, and to leaching by constant water flow for over three years. The Corps reported that modules exhibited stability in all cases; their performance exceeded the performance of other fixation products examined. In our work under EPA sponsorship, we found modules to be stable under extreme thermal shock. They were cycled from a -10°C salt/ice bath to 100°C boiling water, a cycle being of 15 minutes duration with 16 cycles per day over ten days with storage of samples overnight in a freezer. There was no apparent damage to modules and their compressive strengths were at least 95% of their original values.

In further work under EPA sponsorship, we also examined the stability of modules when subjected to harsh leaching solutions such as caustic, hydrogen chloride, citric acid, ammonium hydroxide, ammonium sulfide, and dioxane. In all cases, the modules were found to be stable and exhibit high retention of their hazardous contents. We estimated in our review of state-of-the-art fixation processes that current fixation products would not withstand such harsh leaching solutions. In further leaching studies, we demonstrated the ability of modules to hold 95 weight percent salt under water leaching. In this case, we also speculated that current fixation products would not exhibit such performance when similarly loaded with soluble, inorganic material.

## Apparatus

We are currently completing construction of a prototype apparatus for managing LLRW in commercial-size modules. The apparatus carries out the agglomerating and jacketing process steps; its major components are the two molds which are mounted on a single frame. The apparatus features an indexing table for loading the agglomerate into the jacketing mold. The design criteria for the prototype apparatus include: 1) capability for managing ~300 tons per year of radwaste, 2) transportable, and 3) remotely operable. Thus, the apparatus can function in full-scale LLRW processing demonstrations behind a radiation barrier.

Both the agglomerate and the jacket molds are heated by electrical band heaters and have drilled-in cooling channels for water cooling. A hydraulic system actuates and pressurizes the molds. The agglomerate mold body is a cylindrical steel shell 23-1/2 inches in diameter. The platen fits inside the mold body to pressurize the waste/binder mixture. The mold base is mounted on a linear positioning table that transports the agglomerate to the jacket mold. The jacket mold that fits around the agglomerate is a clamshell mold to facilitate product removal and is capable of being rotated 180° for jacketing by powder molding.

The apparatus is microprocessor controlled for remote operation and the control system is capable

capable of operating the entire process automatically including: heating and pressurizing both molds, regulating cooling water, indexing agglomerate to jacketing mold, rotating the jacket mold, and indexing the module to the unloading platform.

## Cost Benefits

A significant feature of our process is employment of low-cost, mass produced materials. The resins we employ stem from ethylene and butadiene marketed at $0.20 and $0.50 per pound, respectively. These compounds are well known, produced commercially in large amounts, and are used to make polyethylene containers and film, and polybutadiene tires for vehicles. We employ in our jacket fabrication process powdered polyethylene, which is vended to the rotomolding industry for producing holding tanks for fertilizers and harsh chemicals. Powdered polyethylene is presently marketed at approximately $0.45 per pound. Polybutadiene that we employ for fixing contaminants is of the type that is employed in Japan and the U.S. for making corrosion-proof steel frames for automobiles by resin coating them in an electroplating process. The material is offered domestically at about two dollars per pound. By blending the material with polyethylene, we estimate about $1.25 for binder resin. The cost of materials to fabricate a 40 gallon module is estimated to be approximately $40.00. Since foreign sources offer polybutadiene for about 80 cents per pound, potential exists for significantly reducing costs of the material obtained from the domestic market.

The price for polybutadiene is based upon electrodeposition grade material. We estimate a significant portion of its cost lies in its purification for this purpose. Our process, however, does not require a pure material. Consequently, in view of the foreign price and the suitability of cruder polybutadiene for our process, we estimate a significant reduction in cost for this resin is potentially realizable.

In comparison to material costs for other LLRW fixation and containerization schemes, the materials used for managing wastes by Surface Encapsulation are significantly less costly than the materials used in other processes. Table 1 presents a material cost comparison for fixation processes based on cememtitious materials, asphalt, polyester, polybutadiene/polyethylene (Surface Encapsulation), and containerization with High Integrity Containers.

The cost comparison focuses on spent resin solidification. The values used for the waste-to-binder ratios represent widely accepted processing aggregates derived from the technical literature. Costs for High Integrity Containers are based on vendor quotations. The costs are presented on a *dollar* per pound of treated ion-exchange resin.

Table I shows that the Surface Encapsulation Process is the least expensive option for managing spent ion-exchange resins due to the high waste loadings achievable by Surface Encapsulation. Polyester and cementitious solidification systems, and high integrity containerization are significantly more expensive management options. These cost advantages are estimated also to apply to other LLRW such as evaporator bottoms or other ion-exchange media. In addition to material cost advantages, Surface Encapsulation will result in lower transportation and disposal costs due to high waste loadings and, thus, low volume added in the modules.

TABLE I

Comparative Material Costs for LLRW Management

| LLRW FIXATIVE BINDER | BINDER COST $/LB BINDER | FIXATIVE: ION-EXCHANGE RESIN WT. RATIO | COST OF ION-EXCHANGE RESIN MANAGED $/LB RESIN |
|---|---|---|---|
| Cementitious Sodium Silicate | $0.06/lb | 10:1 | $0.60 |
| Asphalt | $0.20/lb | 3:2 | $0.30 |
| Polyester | $0.75/lb | 2:3 | $0.50 |
| Surface Encapsulation | $1.25/lb | 1:10 | $0.12 |
| HIC | $225/55-gal drum | 420 lb/drum<br>55 lb/ft | $0.60 |

## REFERENCES

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