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LEAD MACRO ENCAPSULATION CONCEPTUAL AND EXPERIMENTAL STUDIES - FINAL REPORT (U)

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LEAD MACRO-ENCAPSULATION -
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SUMMARY

Macro-encapsulation, the regulatory treatment for radioactively contaminated lead (mixed) waste has been conceptually and experimentally evaluated for practical application. Epoxy encapsulants molded around lead billets have proven to be exceptionally rugged, easily applied, have high radiation and chemical stability, and minimize required process equipment and production of secondary wastes. This technology can now be considered developed, and can be applied as discussed in this report.

BACKGROUND

The concept of macro-encapsulation for "contaminated/non-reclaimable lead" has evolved from the initial dip/coat application of thermoplastics, such as polyethylene to molding standard lead shapes within a disposable mold with thermosetting epoxy resins. The change in concept was based on the handling difficulties and safety concerns of a heated vat of molten plastic, but most importantly the unavoidable transfer of radioactive contamination from the lead shapes being coated to the entire vat of molten plastic. While no testing with radioactive lead wastes were actually conducted, the presence or absence of radioactivity should have no effect on the testing conducted.

Testing with thermoplastics such as polyethylene immediately demonstrated an inherent process problem: thermoplastics have no melting point, they only become more fluid at higher temperatures. Thus, thermoplastics must be heated sufficiently to allow adequate fluidity for "dip coating" but then the coating drains, slumps, or drips off the lower portions of the object being coated. In experiments using a water quench, strings and streamers of dripping plastic had to be manually removed prior to additional dips or disposal. No feasible application of such resins to irregular shapes was found.

However, if the waste lead were available in uniform shapes, then one could conceivably mold such ingots or bails in a thermoplastic. Encasing a massive ingot or bail in thermoplastics would require molds with standoffs (support points) to achieve the required minimal clearances while minimizing the total required plastic. If the encapsulant were comprised of different materials, then the standoffs could be made of a similar plastic having a much higher softening point, and to which the applied thermoplastic would bond as in a thermo-weld. However, the regulations at present do not permit composite encapsulants, therefore the dimples created by the standoffs would need to be backfilled with hand applied molten resin after the encapsulated billet was demolded.

This conceptualization of a unitized lead waste encapsulated in molds, lead directly to the idea of using thermoset resins, wherein one could pre-mold the required standoffs of the same material as the encapsulant, and thus produce an encapsulated ingot or bail in one molding. Further evolution of this concept produced the process described below and in the following flow diagram.

LEAD ENCAPSULATION PROCESS FLOW DIAGRAM

METHODOLOGY

In brief, the lead is consolidated into a standard sized shape (here after referred to as the billet) just smaller than the dimensions of an inexpensive sheet metal container (open top) to be used as a disposable mold for a thermosetting epoxy resin. The disposable container is prepared to receive the billet by casting and curing a layer of thermosetting resin in the bottom. The billet is then carefully placed within the container and resin is poured into the annulus around and over the top of the billet. After an over night cure the process is complete and final storage/disposal can be carried out. A 5 gallon "lard can", available from SRS stores as a waste container (15 in. diameter x 20 in. high) would serve as a mold for a 400 pound billet.

Consolidation

Lead may be consolidated by either melting/casting or bailing in a hydraulic press. The advantages of pot melting/casting as opposed to bailing is that the equipment is inexpensive, safe and offers the possibility to separate most of the radioactivity in a

slag which could be removed from the surface of the molten metal, collected, "canned" and embedded deep within the cast lead ingot. If handled in this way the surface of the cast lead ingot would have a very low level of transferable activity, thus minimizing health physics concerns. Bailing by contrast requires rather large and expensive equipment to produce high density bails of lead with conceptual diameters of about 15 inches. Moreover, bailing provides no possibility of concentrating the radioactive contamination and reducing the transferable activity at the surface of the billet.

While either process is suitable, melt/casting would appear to be more desirable, especially when it is realized that the melting point for lead is only 328°C, a temperature at which polyethylene was heated to obtain the required low viscosity in the experiments with thermoplastic encapsulation. An electrically heated pot commonly used in construction/plumbing work is adequate for this process. The anticipated rate of production of billets is low enough that an inexpensive reusable split-cylindrical-mold, sized so that its radius and height provide the required regulatory minimum 6mm thick encapsulation.

Choice of thermosetting resins

Various manufactures of thermosetting resins were consulted for recommendations as to the type of resin to be used in this process. Epoxy resins based on aromatic monomers appear to have the most desirable combination of properties. When combined with a silica filler, these resins exhibit exceptional radiation durability. Four different epoxy mixtures from three manufacturers were tested, each having the following desirable properties:

- (1) Thermo-setting at room temperature in <24 hours.
- (2) Working time in excess of 1 hour at room temperature.
- (3) Viscosity <2500 cp.
- (4) Easily mixed epoxy resin and hardener.
- (5) Radiation tolerant through "aromatic" hydrocarbon chemistry combined with high silica loading.
- (6) Non-hazardous/toxic.

Table 1 presents the components and their mixing ratios used to encapsulate 10 pound test ingots. Table 2 gives the MSDS description of the epoxy components listed in table 1.

TABLE 1

Epoxy monomer	Hardener	Mix ratio (wt/wt)	Mix density
Stycast 2651	Stycast 23LV	100/15 (87/13%)	1.50
Thermoset 300	Thermoset 70	100/15	1.50
Epic Resin System S 7151, Part A	Epic Resin System S 7151, Part B	100/20 (83.3/16.7%)	1.43
Epic Resin R-1055	Epic H-5039	100/18 (84.8/15.2%)	1.47

TABLE 2

Component	Chemical Description	Toxicity/Warning
Stycast 2651	filled bisphenol A/ Epichloro hydrin-based epoxy with phenyl glycidyl ether	PEL/TLV limits of 1ppm for the phenyl glycidyl ether but this is <2% in resin; normal ventillation adequate/protect skin and eyes/ do not ingest
Stycast 23LV	aromatic amine catalysts: phenylenediamine and n-methyl pyccolidone	PEL/TLV limits 0.1mg/m3 for the phenylenediamine/nor- mal ventillation adequate/ protect skin and eyes/ do not ingest
Thermoset 300	epoxy resin mixture	low/ protect skin, eyes
Thermoset 70	trioxatridecane diamine polyglycoldiamine mixture	low/ protect skin, eyes, use good ventillation until mixed
Epic S 7151,Part A	modified epoxy resin alkylglycidyl ether	none/protect skin and eyes
Epic S 7151,Part B	modified aliphatic amine: alkyl ether amine, triethanol amine, piperazine, N-aminoethyl piperazine	exposure limits for these amines have not been established/protect skin and eyes
Epic Resin R-1055	modified epoxy resin	none/protect skin and eyes
Epic H-5039	modified aliphatic amine: polyoxyalkyleneamine and polyamide	exposure limits for these amines have not been established/protect skin and eyes

Note: All of the above epoxy resins are approximately 50% silica filled. Since the silica can not become airborne the toxicity limits are meaningless. Should the cured product be machined or ground, respirable silica could then become a concern.

OBSERVATIONS

A test coupon of Thermoset 300/67 epoxy resin was broken into segments and exposed to high intensity gamma radiation (Co^{60} source). After exposures of 10^5 to 10^8 rad single pieces of this were removed for inspection. No visible or physical change in these segments were detected up to and including the maximum exposure. Thus the silica filled epoxy resin has been shown to have the anticipated radiation stability and is totally suitable for encapsulating lead wastes. The test coupon used in this test was made with hardener #67 rather than the #70 chosen because of its viscosity and setting requirements. This change should have no significant effect on radiation stability.

For testing purposes, 10 pound cast ingots were made by casting molted lead into aluminum soft drink cans. The aluminum can was easily peeled off to produce the simulated billet. The simulated casting mold was 28 oz food (baked bean) cans whose dimensions provided the required annular and end thickness of epoxy around the cast billets.

The epoxy resins tested consisted of one very viscous silica filled component and a very fluid hardening agent. Experimentally it was observed that if the hardener were added to the filled resin, it was difficult to disperse the filled resin that wetted the walls of the mixing vessel. However if the reverse order of addition were made the two components would mix well, leaving no uncured resin at the inner wall of the container/mold. This point is important only if the resin is to be mixed in the mold as recommended.

Though not required by regulations, we have evaluated the durability of these epoxy encapsulations to shock and impacts to which they may be subjected before final placement in disposal facilities. This testing with 10 pound cast billets has demonstrated that the silica filled epoxies are much tougher than anticipated. The silica filling greatly decreases the brittleness of the epoxy, apparently by terminating the growth of cracks initiated by impact shock.

RESULTS OF DROPPING TESTS

The planned drop tests using a 0.25 inch butyl rubber as a shock absorbing "sole" attached to the bottom of the encapsulated unit was found to totally protect 10 pound test units dropped through a vertical 6 foot free fall guide tube on to a steel anvil inclined at a 20° angle¹ to the horizontal. The 60 ft.lb. force generated by this drop is absorbed by the butyl rubber, over an area described as a chord across the bottom edge of the test unit. This rate of energy absorption is greater than that generated by a

¹ The 20° angle simulates the maximum anticipated rotation a production unit could undergo when dropped 4 inches from a vertical position. Striking a massive surface at such an angle produces a near maximum shock to the epoxy encapsulant by the available forces generated by free fall.

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400 pound (5 gallon) unit dropped from 3-4 inches (100-120 ft.lb.) due to rough handling by a fork lift operator. To facilitate observation of shock damage the encapsulated units were removed from the "28 ounce" mold prior to the drop tests. What ever protection such a mold might provide has not been tested but should not be significant.

After passing the maximum (6 ft) drop with the rubber shock absorbers, the tests were repeated without any rubber pad, and again the 10 pound unit suffered only the slightest edge spalling up to about 30 ft.lbs. and only slight bulging (mushrooming) at the impact point for higher drops (up to the 6 foot maximum tested).

The effectiveness of the rubber pad (attached to the bottom of the epoxy encapsulated lead billet) in absorbing shock could not be quantized since the epoxy did not fail even the greatest free fall allowed for in these tests. However, it was quite clear that with the rubber sole the edges of the test ingot were totally protected even though the rubber was permanently "dented" by the greater impacts. While the addition of rubber protective "soles" to encapsulated units does not appear to be required for the 3-4 inch inadvertent drops simulated in these tests, such absorbers would add additional ruggedness to the waste form if added.

CONCLUSIONS

The process technology tested and reported herein, has been shown to be a practical approach to the macro-encapsulation of contaminated lead. It meets all regulatory requirements to date, can be easily implemented, and will thus be recommended for application at the Savannah River Site and elsewhere in the DOE complex.

QA STATEMENT

The data and observations upon which this report is based, were collected under the guidance of the Task Technical and QA Plan² and referenced procedures, and recorded in a laboratory notebook, WSRC-NB-92-163, pp 42-51 which acts as the primary repository for all data of this study.

² E. G. Orebaugh, "Macro-encapsulation, A Treatment for Radioactive Lead Waste..(U)", WSRC-RP-92-477 & 489, (Task Technical and QA Plan)

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