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Treatment of waste printed wire boards in electronic waste for safe disposal

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Abstract

The printed wire boards (PWBs) in electronic waste (E-waste) have been found to contain large amounts of toxic substances. Studies have concluded that the waste PWBs are hazardous wastes because they fails the toxicity characteristic leaching procedure (TCLP) test with high level of lead (Pb) leaching out. In this study, two treatment methods – high-pressure compaction and cement solidification – were explored for rendering the PWBs into non-hazardous forms so that they may be safely disposed or used. The high-pressure compaction method could turn the PWBs into high-density compacts with significant volume reduction, but the impact resistance of the compacts was too low to keep them intact in the environment for a long run. In contrast, the cement solidification could turn the PWBs into strong monoliths with high impact resistance and relatively high compressive strength. The leaching of the toxic heavy metal Pb from the solidified samples was evaluated by both a dynamic leaching test and the TCLP test. The dynamic leaching results revealed that Pb could be effectively confined in the solidified products under very harsh environmental conditions. The TCLP test results showed that the leaching level of Pb was far below the regulatory level of 5 mg/L, suggesting that the solidified PWBs are no longer hazardous. It was concluded that the cement solidification is an effective way to render the waste PWBs into environmentally benign forms so that they can be disposed of as ordinary solid wastes or beneficially used in the place of concrete in some applications.

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1. Introduction

While the use of electronic products continues to increase worldwide, the life spans become shorter and shorter due to the rapid advances in technology, leading to an increasing generation of electronic waste (E-waste). The U.S. Environmental Protection Agency (EPA) has found that E-waste components already constitute 1% of the municipal solid waste (MSW) streams [1] and they are growing 2–3 times faster than any other components in MSW [2]. It is estimated that as many as 500 million personal computers (PCs) will become obsolete and enter the MSW stream between 2000 and 2007 [2]. Electronic products contain a complex array of toxic substances of which some can readily leach out in certain environment settings. Among all the components in electronic products, the printed wired boards

0304-3894/\$ - see front matter © 2006 Elsevier B.V. All rights reserved. doi:10.1016/j.jhazmat.2006.11.039 (PWBs) contain the most toxicants in both quantity and variety. The most significant toxicants in PWBs are the heavy metal lead (Pb) and the brominated flame retardants (BFRs). The previous toxicity characteristic study of the PWBs in PCs by the EPA toxicity characteristic leaching procedure (TCLP) has shown that Pb concentrations in the TCLP extracts of the vast majority of the PWBs ranged from 150 to 500 mg/L, which are 30–100 times the regulatory level of 5 mg/L for classifying a waste as hazardous [3].

The ideal solution to the waste PWBs would be recycling and reuse, but this is currently not economically viable. The metallurgical processes required for the recovery of the noble metals are very sophisticated. The costs for processing and for dealing with the secondary pollutants generated in the processes surpass the market values of the materials that could be recovered from the PWBs. At present, landfilling remains to be the predominant way for the disposal of the E-waste including PWBs. Realizing that Pb leaches out from PWBs at much greater level than the toxicity characteristic limit, concerns on the disposal

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of PWBs in landfills continue to grow. Landfill bans on E-waste that contains PWBs have been planned in some states in the U.S. [4]. On the other hand, disposing E-waste as hazardous waste in hazardous waste landfills is not generally practical because (1) hazardous waste landfills are not available in all places; long-distance transportation is needed in many areas; (2) hazardous waste disposal is much more costly than ordinary waste; and (3) the number and space of hazardous waste landfills are limited; if much of the E-waste goes to hazardous waste landfills, the hazardous landfill space will be in short, making the disposal of the conventional hazardous waste difficult. Co-disposal of the E-waste with ordinary MSW in sanitary landfills can be a promising solution if the E-waste is properly treated and rendered into non-hazardous forms in an economical way. In this study, two methods – high-pressure compaction and cement solidification - were explored for the treatment of PWBs so that they may be disposed in ordinary landfills without posing potential hazards to the environment. The effectiveness of the treatment in reducing the environmental hazards was evaluated by a dynamic leaching test and a slightly modified TCLP test.

2. Materials and methods

2.1. Materials

PWB samples were collected from the Computer Recycling Project at Jackson State University, Mississippi. The PWBs included motherboards and expansion cards from desktop PCs with a wide range of models and manufacturers. The large metal parts of the PWBs such as the radiators on the motherboards and the steel pieces for connecting to the PC frames were removed. The PWBs were then shredded by a shear-type shredder (Amerishred, AMS 7500 Series). The PWBs were passed through the shredder twice. They were shredded into 19-mm wide strips the first time and then into approximately square particles by feeding the strips perpendicular to the shear discs of the shredder. A small number of particles that were still longer than 19 mm after the second shredding were further cut into the ones no bigger than 19 mm by a hand shear. A picture of the shredded PWBs is shown in Fig. 1(a).

Two types of cement were used as binders for the solidification of the shredded PWBs. One was the most commonly available Portland cement (Type I, Ash Grove Company). The low cost and widespread availability of the raw materials (limestone, shales, and other naturally occurring materials) for producing the Portland cement make it one of the lowest cost construction materials throughout the world. It is also one of the most commonly used binders for the solidification and stabilization of hazardous waste. Another type of cement used in the study was slag cement. The slag cement is also called ground granulated blast furnace slag. It is a byproduct of steel industry, produced during the reduction of iron ore to iron in a blast furnace. It has been used for many years as a supplementary cementitious material in Portland cement concrete, either as a mineral admixture or as a component of blended cement [5]. Its slow-setting property sometimes limited the extent of its use in concrete. In stabilization of waste, this is not typically an issue. The slag cement used in this study was provided by Holcim (US) Inc., Birmingham, Alabama.

Sand (QUIKRETE All-Purpose Sand) and gravel (QUIK-RETE All-Purpose Gravel) were used as fine and coarse aggregates, respectively, in the solidification. They both met the specifications for fine and coarse aggregates as specified in the standard specification ASTM C-33 of American Society of Testing and Materials [6].

2.2. High-pressure compaction

The high-pressure compaction technology developed by the Capsule Pipeline Research Center at the University of Missouri, Columbia was used to compact the PWBs. The unique high pressure (up to 200 MPa) of this technology could turn many types of waste materials into dense and useful products [7]. It was intended that this technology could convert the PWBs into dense, strong, and almost-impermeable solid monoliths/compacts so that the volume could be significantly reduced and leaching of toxic substances largely minimized. The compacts may be co-disposed with municipal solid waste in the ordinary landfills if the compacts meet certain criteria.

The shredded PWBs were compacted at five different pressures 69, 103, 138, 172, and 207 MPa. The quality of the compacts was evaluated by density and the impact resistance as described in Section 2.5.

2.3. Cement solidification

The Portland cement and the slag cement were used as the main binders to produce solidified PWB specimens. The materials in the specimens included cement (either Portland or slag), sand, shredded PWB, and water. The shredded PWB played a role as the coarse aggregate in concrete. To achieve the maximum inclusion of PWB and minimum use of cement, the ratios among these materials were optimized by trial-and-error. The final ratios (weight basis) used were water/cement 1.0, sand/cement 2.5, and PWB/cement 1.5. The procedure for making the specimens was in compliance with the ASTM C-192 standard practice for making concrete test specimens [8] except that the shredded PWB was used in the place of the coarse aggregate. The cement and sand were first mixed thoroughly by hand in a stainless steel bowl. Then the shredded PWB was added and mixed. The water was added the last and mixed until the mortar is homogeneous in appearance and ready for molding. The molds were made of high-density polyethylene (HDPE) with an inner diameter of 76.2 mm and a height of 152 mm. The mortar was placed in the mold using a scoop in layers of 50 mm and each layer was rodded by hand for a minimum of 25 strokes. After the molds were filled, they were capped by a plastic cover that could maintain the mold water-tight for setting. The setting period was 24 ± 8 h for the Portland cement specimen and $72 \pm 8 h$ for the slag cement specimens because the setting for the latter was much slower. After the setting periods, the molds were removed and the cylindrical specimens





Fig. 1. Pictures of (a) shredded PWB, (b) compacted PWB at 103 MPa, (c) cement-solidified PWB specimen, and (d) inside view of a solidified PWB specimen.

were continually cured at the room temperature $(22 \pm 2 \,^{\circ}C)$ in closed plastic bags where the relative humidity was maintained at above 95%. The curing time was 7 days for the specimens to be used for dynamic leaching test and 28 days for those to be used for compressive strength test. For comparison purpose, blank specimens were made. There were no PWB in the blanks and gravel was used as the coarse aggregate. The water/cement and sand/cement ratios in the blanks were the same as in the PWB-containing specimens, but a higher gravel/cement ratio of 1.7 was used for the blanks in order to achieve the same volume fraction as PWB in the specimens. The procedure for producing the blanks was the same as for the PWB-containing specimens. Pictures of a cement-solidified PWB specimen and an inside view of the specimen are shown in Fig. 1(c) and (d), respectively.

2.4. Compressive strength test

The compressive strengths of the solidified specimens that had been cured for 28 days were tested. The specimens included blanks of both Portland cement and slag cement in triplicates and PWB-containing specimens solidified by both Portland cement and slag cement in triplicates. The test was performed according to the standard method ASTM C-39 [9].

2.5. Drop test

The impact resistances of the compacts made from the high-pressure compaction and the cylinders from the cement solidification were evaluated by a drop test. This test was an adaptation to the ASTM method D-440, a drop shatter test for coal [10]. The samples were dropped twice onto a concrete floor from 1.83 m. An impact resistance index (IRI) introduced by Richards [11] was used to evaluate the impact resistance of the samples. The IRI was calculated as $IRI = (100 \times N)/n$, where N was the number of drops, and n was the total number of pieces after N drops. Since the samples were dropped for two times in the test, N was always 2 and the maximum value of IRI would be 200 when the sample remained unbroken after two drops. If the sample broke into 10 pieces, the IRI would be 20. A result of IRI being smaller than 20 indicates a very poor impact resistance. Note that after the first drop, all the pieces that were less than 5% of the initial weight of the samples were not used in the second drop and all the pieces smaller than 5% of the initial weight after the second drop were not included in the calculation of IRI.

2.6. Dynamic leaching test

The effect of solidification on reducing or preventing the leaching of contaminants from PWBs was evaluated by a

dynamic leaching test (DLT). Two types of leaching fluids, TCLP extraction fluid #1 [12] and synthetic precipitation leaching procedure (SPLP) extraction fluid #1 [13], were used in the test. The former was a buffer solution consisting of 5.7 mL of glacial acetic acid and 64.3 mL of 1N NaOH solution per liter. The pH was 4.93 ± 0.05 . The latter was made by adding 60/40 wt.% mixture of sulfuric and nitric acids with suitable dilution to reagent water until the pH is 4.20 ± 0.05 . The TCLP was designed to simulate the worst case leaching scenario in MSW landfills. The SPLP was designed to evaluate the leachability of contaminants from soil and waste samples caused by rainwater of low pH. The SPLP extraction fluid #1 is to be used for soil samples for sites that are east of Mississippi River and solid and liquid wastes.

For each extraction fluid, four solidified specimens were tested: one blank of Portland cement, one blank of slag cement, one Portland-cement-solidified PWB specimens, and one slag-cement-solidified PWB specimen. The specimens were submerged in the extraction fluids with a liquid-to-solid ratio of 4:1 on weight basis in HDPE containers. The containers were capped, but not air-tight. The total leaching period for all the specimens was 61 days. Leaching cycles of 1, 3, and 7 days were used for the first 11 days and a cycle of 10 days for the rest leaching periods. At the end of each cycle, a liquid sample (DLT leachant) was taken and the extraction fluid was completely renewed by the fresh one to start another cycle. The pH of DLT leachant was measured first, then the pH was adjusted to between 1.5 and 2.0 by concentrated nitric acid for the analysis of Pb later. Lead was chosen because it was the only heavy metal that could leach out in significant amounts in the TCLP test of the PWBs and cause the PWBs to be hazardous waste [3]. An atomic absorption spectrophotometer (Shimadzu AA-6200) was used to analyze the Pb concentration according to EPA Method 7000A [14].

2.7. TCLP test

The solidified PWB-containing specimens that had been tested for compressive strengths as stated in Section 2.4 were further crushed into particles with a maximum size of some 19 mm, which was equal to the largest size of the shredded PWBs in the specimens. The crushed samples were dried at 105 ± 5 °C and then tested according to the standard TCLP method except that the sizes of the particles were not further reduced to 9.5 mm. Representative samples of 100 g were taken and mixed with the TCLP extraction fluid #1 at a liquid-to-solid ratio of 20:1 on



Fig. 2. Effect of compaction pressure on the density of the PWB compacts.

weight basis in a HDPE bottles, then agitated in a rotary extractor at 30 rpm and 22 ± 2 °C for a period of 18 ± 2 h. After the agitation, the samples were filtered through a 0.6–0.8 µm glass-fiber filter. The filtrate was analyzed for Pb concentration using the same method as for the DLT leachant.

3. Results and discussion

3.1. Physical properties of treatment products

The densities of the PWB compacts produced at different compaction pressures are shown in Fig. 2. The density of the compacts increased significantly with the increase of the compaction pressure for pressures lower than 100 MPa; the increase was about 0.013 g/cm³ per MPa. For pressures higher than 100 MPa, the increase became small, only 0.0014 g/cm^3 per MPa on the average. The compacts made at pressures lower than 100 MPa were very weak; pieces from the surface could easily fall off by hand touching. The compacts made at pressures higher than 100 MPa had a good integrity. A picture of the compact made at 103 MPa is shown in Fig. 1(b). The impact resistances of all the compacts were very low. The IRIs of all the compacts made at pressures from 69 to 207 MPa were smaller than 20. This unexpected shortfall of the products makes the application of the high-pressure compaction technology the treatment of waste PWBs very limited.

The physical properties of the cement-solidified products (cylinders) including the density, compressive strength, and impact resistance are given in Table 1. These properties were measured after the specimens had been moist-cured for 28 days.

Table 1
Physical properties of cement-solidified specimens

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Cement type	Specimen	Density (g/cm ³)	Compressive Strength (MPa)	IRI ^a
Slag cement	Blank Solidified PWB	$\begin{array}{c} 2.04 \pm 0.01^{\rm b} \\ 1.96 \pm 0.02 \end{array}$	$\begin{array}{l} 8.31 \pm 0.09^{\rm b} \\ 4.89 \pm 0.32 \end{array}$	200 200
Portland cement	Blank Solidified PWB	$\begin{array}{c} 2.08 \pm 0.04 \\ 2.01 \pm 0.01 \end{array}$	$\begin{array}{c} 13.7 \pm 1.3 \\ 7.93 \pm 0.65 \end{array}$	200 200

^a Impact resistance index (see Section 2.5).

^b Standard deviation of triplicate samples.

While the densities of the Portland cement products were slightly higher than those of the slag cement ones, the compressive strengths of the former were much higher than the latter. For the products with the same type of cement, the blanks had much higher compressive strength than the PWB-containing ones. In the case of the Portland cement, the average compressive strength was 14 MPa for the blanks and 7.9 MPa for the PWB-containing ones. This indicates that the adhesion between the cement and shredded PWB is weaker than that between the cement and the gravel. The blanks were made the same way as for the normal concrete and the compressive strength matched the value of the ordinary concrete. The compressive strength for concrete is highly dependent on the water/cement ratio-the lower the ratio is, the higher compressive strength the concrete will have. The average 28-day strengths of Portland cement concrete are 15–45 MPa for water/cement ratios from 0.79 to 0.37, respectively [15]. Since the water/cement ratio used in this study was 1.0 for all the specimens, the compressive strength of the blanks was at the lower end of the average range. It is expected that the strengths of both the blanks and the PWB-containing specimens would increase as the water/cement ratio decreases. Even with the 1.0 ratio, all the products are strong enough for the purpose of co-disposal with MSW and some uses which do not require high compressive strength. The slag-cement-solidified products had lower compressive strengths than the Portlandcement-solidified ones, 8.3 MPa for the blanks and 4.9 MPa for the PWB-containing ones. They are still sufficiently strong for disposal purpose.

The impact resistances of all the cement-solidified products were very high. They remained solid in the drop test. Some specimens only had small pieces chipped of the edges, but no weight loss of greater than 1% of the specimens was observed. Therefore, the IRIs for all the samples were 200 which is the maximum and indicates high impact resistance.

3.2. Volume and weight changes

The compaction could achieve high volume reduction of the shredded PWB. A volume reduction ratio was calculated by the ratio of the density of the compact to the bulk density of the shredded PWB. The bulk density of the shredded PWB was measured by placing them in a container and shaking to the smallest achievable volume, then dividing the weight of the PWB by the volume it occupied. The bulk density of the shredded PWB was measured to be 0.643 g/cm³. Therefore, the volume reduction ratios were 1.8, 2.5, 2.6, 2.7, and 2.7 for the compaction pressures 69, 103, 138, 172, and 207 MPa, respectively. There was no weight change in compaction because no other materials were involved in the treatment.

The solidification increased the bulk volume of the shredded PWB slightly although the weight increase was significant. The ratio of the volume of a solidified specimen to the bulk volume of the shredded PWB solidified in the specimen was 1.14, an increase of 14% in volume. The average ratio of the weight of the solidified specimens to the weight of the shredded PWB solidified in the specimens was 3.55, an increase of 255% in weight. This is an inevitable drawback of the solidification treatment. Since the materials used are inexpensive, the costs may be offset by the benefits from the avoided hazardous treatment or disposal costs or beneficial use of the solidified products.

3.3. Dynamic leaching of Pb

The pH and Pb concentrations of the DLT leachant at the end of each leaching cycle were measured for the blanks and the solidified PWB specimens of both Portland cement and slag cement in two different extraction fluids. The results are given in Tables 2 and 3, respectively. For the Portland cement specimens in both the SPLP and TCLP extraction fluids, no Pb was detected in the leachants for all the leaching cycles. For the slag cement specimens, Pb was detected in the leachant of SPLP extraction fluid in only three leaching cycles (cycles 2, 3, and 4) at very low levels (0.11, 0.92, and 0.19 mg/L, respectively). No Pb was detected in the leachant of TCLP extraction fluid. The leaching of Pb from slag cement specimen in the SPLP extraction fluid was due to the fact that a few PWB pieces exposed on the surface of the specimen. The Pb in the exposed pieces dissolved. After three leaching cycles, the most of the Pb that was directly in contact with the leaching fluid dissolved away and the leaching ceased. In contrast, no PWB pieces were seen on the surfaces of all other specimens in the test. Therefore, no detectable Pb was found. The pH of SPLP extraction fluid increased dramatically for both

Table 2

Lead concentration and pH at the end of each leaching cycle in DLT test of Portland-cement-solidified PWB specimens

Leaching cycle	Leaching period (day)	SPLP extraction fluid		TCLP extraction fluid			
		pH for blank	pH for solidified PWB	Pb concentration ^a (mg/L)	pH for blank	pH for solidified PWB	Pb concentration ^a (mg/L)
1	1	11.3	11.8	ND ^b	5.5	5.5	ND ^b
2	3	11.9	11.4	ND	5.4	5.4	ND
3	7	11.6	11.0	ND	5.4	5.5	ND
4	10	11.3	11.2	ND	5.3	5.4	ND
5	10	11.1	10.7	ND	5.5	5.5	ND
6	10	10.1	10.0	ND	5.7	5.6	ND
7	10	9.6	9.6	ND	5.4	5.5	ND
8	10	9.6	9.1	ND	5.3	5.4	ND

^a Pb concentrations in this column are for the DLT leachants of solidified PWB specimens. The Pb concentrations for all the blanks were not detectable. ^b Not detectable. X. Niu, Y. Li / Journal of Hazardous Materials 145 (2007) 410-416

Lead concentration and pH at the end of each leaching cycle in DL1 test of stag-cement-solidined P wb specificities							
Leaching cycle	Leaching period (day)	SPLP extraction fluid		TCLP extraction fluid			
		pH for blank	pH for solidified PWB	Pb concentration ^a (mg/L)	pH for blank	pH for solidified PWB	Pb concentration ^a (mg/L)
1	1	10.1	10.3	ND ^b	5.2	5.3	ND ^b
2	3	10.1	10.1	0.11	5.1	5.2	ND
3	7	9.6	9.4	0.92	5.3	5.5	ND
4	10	8.8	9.3	0.19	5.4	5.5	ND
5	10	9.0	8.1	ND	5.3	5.4	ND
6	10	8.7	8.6	ND	5.3	5.3	ND
7	10	8.2	8.2	ND	5.4	5.5	ND
8	10	7.6	7.8	ND	5.4	5.4	ND

Lead concentration and pH at the end of each leaching cycle in DLT test of slag-cement-solidified PWB specimens

^a Pb concentrations in this column are for the DLT leachants of solidified PWB specimens. The Pb concentrations for all the blanks were not detectable. ^b Not detectable.

the Portland cement and slag cement specimens, from 4.2 to 10 for slag cement specimens and to almost 12 for Portland cement specimens at the ends of the first a few leaching cycles. The increase diminished as the leaching cycles proceeded because the alkalinity of the cement was gradually neutralized by the acidic leaching fluid.

The pH of TCLP extraction fluid increased slightly for both the Portland cement and slag cement specimens, from 4.93 to 5.3 for slag cement specimens and to 5.4 for Portland cement specimens for all the leaching cycles. This was due to that TCLP extraction fluid is a strong buffer solution that could stabilize the pH. It was also observed that there was no significant difference in the pH values of the leachants of the blank and the PWBcontaining specimen in the same extraction fluid (either SPLP or TCLP). This suggested that the pH was dominated by the cements.

Although the pH of the TCLP extraction fluid at the end of each leaching cycle for both the Portland cement and slag cement specimens was slightly acidic, there was no detectable amount of Pb leached out. Therefore, both the cements had strong ability to retain Pb under harsh (acidic) environmental conditions. After 61 days of leaching test, all the specimens remained strong and integral. No physical change could be observed. The low permeability of the concrete and the mineralization and absorption of Pb by the cement contribute to the prevention of Pb from leaching out.

3.4. TCLP leaching of Pb

Table 3

No detectable amount of Pb was found in the TCLP extracts of the slag-cement-solidified PWB samples and very low level of Pb (0.19 \pm 0.07 mg/L) was detected in the TCLP extracts of the Portland-cement-solidified PWB samples. This was in a sharp contrast to the TCLP results for PWB alone, which the Pb concentration in the TCLP extracts was 150–500 mg/L [3]. This suggests that even in the worst case scenario in landfills, i.e., the cement-solidified products were completely crushed and the landfill leachate were at its lowest possible pH, the leaching of Pb could still be prevented to a minimum level. A recent study on the factors affecting TCLP leachability of Pb from PCs [16] showed that the particle size of PWB samples has little effect on the leaching of Pb when the particle sizes are near 9.5 mm, which is the maximum size required by the standard TCLP test. This implies that TCLP test results would not have been different had the size of the particles been reduced to a maximum of 9.5 mm instead of 19 mm. Thus, the solidified PWBs are no longer hazardous waste because the Pb concentration in TCLP extract is far below 5 mg/L, which is the regulatory level to determine whether a solid waste is hazardous or not. While the Portland cement was sufficiently effective in reducing the leaching of Pb to more than one order of magnitude lower than the regulatory level in the TCLP test, the slag cement was more effective so that no Pb above the detection limit (0.1 mg/L)was detected. Since fast setting is not essential in the solidification treatment of wastes, the slow-setting property of the slag cement does not downgrade its application in the solidification of waste PWBs.

4. Conclusion

The high-pressure compaction method can turn the waste PWBs into high-density compacts with significant volume reduction, but impact resistance of the products is so low that its application in the treatment of PWBs is limited. The cement solidification was found to be an effective way to turn the PWBs into strong monoliths with high compressive strengths and impact resistance. Both Portland cement and slag cement showed high effectiveness in this treatment. The DLT test of the cement-solidified PWB specimens showed that the leaching of the most concerned toxic heavy metal Pb in the PWBs can be effectively prevented even under harsh (acidic) environmental conditions. The TCLP test of the cement-solidified PWB specimens resulted in a Pb concentration in the TCLP extract far below the regulatory level of 5 mg/L, suggesting that the solidified PWBs are no longer hazardous. They can be safely co-disposed with MSW in ordinary landfills. The solidified products may also be beneficially used in the place of concrete where high compressive strength is not critical. The cost of the solidification treatment may be justified by the avoided hazardous waste disposal cost, long-term environmental benefits, and/or beneficial use of the solidified, concrete-like products.

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