

A microencapsulation process of liquid mercury by sulfur polymer stabilization/solidification technology. Part II: Durability of materials⁽¹⁾

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Abstract

Under the European LIFE Program a microencapsulation process was developed for liquid mercury using Sulfur Polymer Stabilization/Solidification (SPSS) technology, obtaining a stable concrete-like sulfur matrix that allows the immobilization of mercury for long-term storage. The process description and characterization of the materials obtained were detailed in Part I. The present document, Part II, reports the results of different tests carried out to determine the durability of Hg-S concrete samples with very high mercury content (up to 30 % w/w). Different UNE and RILEM standard test methods were applied, such as capillary water absorption, low pressure water permeability, alkali/acid resistance, salt mist aging, freeze-thaw resistance and fire performance. The samples exhibited no capillarity and their resistance in both alkaline and acid media was very high. They also showed good resistance to very aggressive environments such as spray salt mist, freeze-thaw and dry-wet. The fire hazard of samples at low heat output was negligible.

Keywords

Mercury; Sulfur polymer stabilization/solidification (SPSS); Durability; Water absorption; Salt aging; Freeze-thaw.

Proceso de microencapsulación de mercurio líquido mediante tecnología de estabilización/solidificación con azufre polimérico. Parte II: Durabilidad de los materiales

Resumen

Dentro del Programa Europeo LIFE, se ha desarrollado un proceso de microencapsulación de mercurio líquido, utilizando la tecnología de estabilización/solidificación con azufre polimérico (SPSS). Como resultado se ha obtenido un material estable tipo concreto que permite la inmovilización de mercurio y su almacenamiento a largo plazo. La descripción del proceso y la caracterización de los materiales obtenidos, denominados concretos Hg-S, se detallan en la Parte I. El presente trabajo, Parte II, incluye los resultados de los diferentes ensayos realizados para determinar la durabilidad de las muestras de concreto Hg-S con un contenido de mercurio de hasta el 30 %. Se han utilizado diferentes métodos de ensayo estándar, UNE y RILEM, para determinar propiedades como la absorción de agua por capilaridad, la permeabilidad de agua a baja presión, la resistencia a álcali y ácido, el comportamiento en niebla salina, y la resistencia al hielo-deshielo y al fuego. Las muestras presentaron una capilaridad extremadamente baja y una resistencia alta tanto a álcali como a ácido. También mostraron una elevada resistencia en condiciones ambientales muy agresivas como niebla salina, hielo-deshielo y humedad-sequedad.

Palabras clave

Mercurio; Estabilización/solidificación con azufre polimérico (SPSS); Durabilidad; Absorción de agua; Niebla salina; Hielo-deshielo.

1. INTRODUCTION

The United Nations Environment Program (UNEP) established the Mercury Program with the aim of delivering activities on mercury and to support negotiations of an international instrument for the control of mercury. Thus a series of Governing Council Decisions have been established since 2001

which include the prevention, minimization and management of mercury waste and also its long-term storage and disposal^[1]. European Directive 2000/60/EC considers mercury to be a priority hazardous substance due to its adverse effects on human health and the environment^[2]. EU Council and European Parliament (Regulation 1102/2008-EC)^[3] set 2011 as the cutoff date for industrial use of

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Hg and the banning of mercury exports, and addresses the issue of safe storage of metallic mercury. As a result all excess Hg must be stored in safe conditions in secure places until definitive stabilization policies are established. Thus there is a need for a process that effectively stabilizes elemental Hg, preferably using inexpensive additives and/or which can be economically operated at both small and large scales^[4].

Solidification/stabilization (S/S) is accepted as a well-established disposal technique for the treatment of hazardous waste. The effectiveness of S/S products is basically defined by two parameters: strength and leach resistance. The former is an indicator of solidification and the second assesses the extent of fixation^[5 and 6]. The continuing need to develop improved and more economical waste management techniques has raised the potential importance of solidification technology throughout the world. However, conventional S/S technologies based on cement-like materials cannot effectively reduce the leachability of mercury as it tends to hydrolyze to form mercuric oxide, thus maintaining a strong potential to volatilize^[7]. Accordingly, a number of S/S based technologies have been developed looking for alternatives to cement. Sulfur polymer solidification stabilization technologies (SPSS) are being tested for using in the long-term storage of hazardous waste in general and mercury in particular^[8-10]. These processes are based on the use of a modified sulfur, which has less shrinkage and hence develops less residual stress upon cooling; on the one hand because it remains in the monoclinic modification of S_{β} and does not undergo a phase transformation to the orthorhombic form of S_{α} upon solidification, and on the other because its thermal expansion coefficient is much lower than that of unmodified S_{α} . The modified sulfur is often incorrectly referred to as sulfur polymer cement (SPC). Its use yields composite materials with high strength, low water absorption, and high resistance to acid and basic environments^[11].

In Part I^[12], described the development of a batch-type microencapsulation process for liquid mercury from the chloralkali industry using SPSS technology. This process allows the immobilization of high amounts of mercury (up to 30 %), rendering concrete-like solid monoliths as the end product. This product exhibits excellent mechanical properties (compressive strength 53-61 MPa and flexural strength 7-10 MPa) and the toxicity characteristic leaching test reveals a mercury concentration in leachates well below the 0.2 mg L⁻¹ set out in US EPA Land Disposal Restrictions (LDRs). Mechanical properties and leachability are the factors commonly studied in the bibliography to

test the effectiveness of stabilization processes. In Part II the authors have tested the behavior of the Hg-S concrete samples, obtained in Part I, in a number of different aggressive environments, e.g. in salt mist, freeze-thaw and wet-dry situations, alkali/acid media, etc., with a view to more fully assessing the effectiveness of the stabilization process and assuring the durability of the monoliths in long-term storage. Besides, properties such as capillary water absorption and low pressure water permeability were also studied. This study, like that described in Part I^[12], was performed within the European Project MERSADE (LIFE06 ENV/ES/PRE/03).

2. EXPERIMENTAL

2.1. Materials

The materials considered in this study are 40x40x160 cm Hg-S concrete samples obtained by the SPSS process reported in Part I^[12]. To achieve an end product with concrete-like characteristics, commercial gravel (< 6.3 mm) and siliceous sand (< 4 mm) were used as inert aggregate materials and CaCO₃ (< 0.125 mm) as filler, at a constant gravel/sand/filler ratio of 30/60/10 wt/wt/wt for all the samples. Mixtures of metacinnabar, aggregates and filler were heated to 145 °C and stirred for 30 min. Six solid blocks of around 800 g in weight were obtained per batch. The liquid mercury content in the mixture was 20 and 30 % mass percentage, which corresponds to a HgS percentage ranging between 23.2 - 34.0 %. The monolithic samples obtained are referred to as Hg20SC and Hg30SC, and the mercury-free sulfur-concrete reference sample as SC.

2.2. Methods

The durability of a concrete structure is closely associated to the permeability of its surface layer, which is responsible for limiting the entry of substances that can initiate or propagate possible deleterious actions (water, oxygen, alkalis, salts, etc.). The behavior of the samples was studied in different environments by the application of various test methods to determine their physical properties (water absorption and permeability) and durability (aging test, freeze-thaw and wet-dry resistance, alkaline and different pH solution resistance, and fire performance).

2.2.1. Physical properties: Water absorption

Capillary water absorption and the water absorption coefficient (W_s) were determined according to standard UNE-EN 480-5:2006^[13]. As shown in figure 1, the samples were placed vertically inside a sealed chamber (55 % relative humidity) with standing water (5 cm deep). After 10 min, 30 min, 1, 3, 6, 24, 72 h and 28 days of contact with the water the samples were dried with paper to remove any excess water and then weighed. Mass variation (ΔM) and capillary absorption coefficient (W_s) values were calculated using equation (1):

$$W_s = \Delta M/A = \frac{M_j - M_o}{16} \quad (1)$$

where M_j is the sample mass after 28 days, M_o the initial mass, and 16 cm² the sample area (4x4 cm).

2.2.2. Physical properties: Water permeability

Water permeability under low pressure was determined using the RILEM method (Reunion Internationale des Laboratoires d'Essais et de



Figure 1. Positioning of sample in chamber for capillary absorption test.

Figure 1. Posicionamiento de la muestra en el interior de la cámara para el ensayo de absorción por capilaridad.

Recherches sur les Materiaux et des Constructions/ International Union of Testing and Research Laboratories for Materials and Structures)^[14]. The method determines the amount of water absorbed per surface area unit when the sample is subjected to a pressure equivalent to a 10 cm water column per unit of time. The aim is to reproduce the real pressure of rain drops at high speed (approx. 140 km h⁻¹) against an outer wall. Figure 2 shows the experimental setup for the horizontal application of the Rilem Test Method. To measure the amount of water absorbed by the surface in a certain time, a set of small pipe-shaped graduated tubes are fixed to the wall zones to be studied. The amount of absorbed water is indicated by the drop in the water level observed in the tubes. Measurements were carried out at 2, 5, 10, 15, 30, 60 and 180 min.

2.2.3. Durability: Alkali resistance

Alkali resistance for hydrophobic impregnations has been determined according to standard UNE-EN 13580:2003^[15]. The monolithic samples were placed in individual 1550 ml volume beakers containing a 5.6 g L⁻¹ KOH solution which covered the samples. The beakers were covered with transparent film for (21.0 ± 0.1) days. Tests were performed in a properly conditioned curing chamber. The samples were then air-dried until their masses were equal to the initial mass ± 2 g. A second immersion test was carried out and the mass increase after alkali treatment was determined ($i_{t(alk)}$). The absorption coefficient (AR_{alk}), expressed as % mass (g), was calculated using equation (2)

$$AR_{alk} = \frac{i_{(alk)}}{i_i} \times 100 \quad (2)$$

where i_i is the mass of the sample before immersion in the alkaline solution and $i_{t(alk)}$ is the mass after its second immersion.

This same methodology was used to determine the water absorption and resistance to aqueous solutions of different pH values, in this case using commercial buffer solutions with pH values of 2, 4, 6, 8 and 10 instead of the KOH solution defined in the standard test^[15]. Tests were performed for a series of 4x4x16 cm samples with the higher mercury content (Hg30SC). As a pH value of 2 is the most aggressive for this kind of material, the test at this pH was also performed with the reference sample.

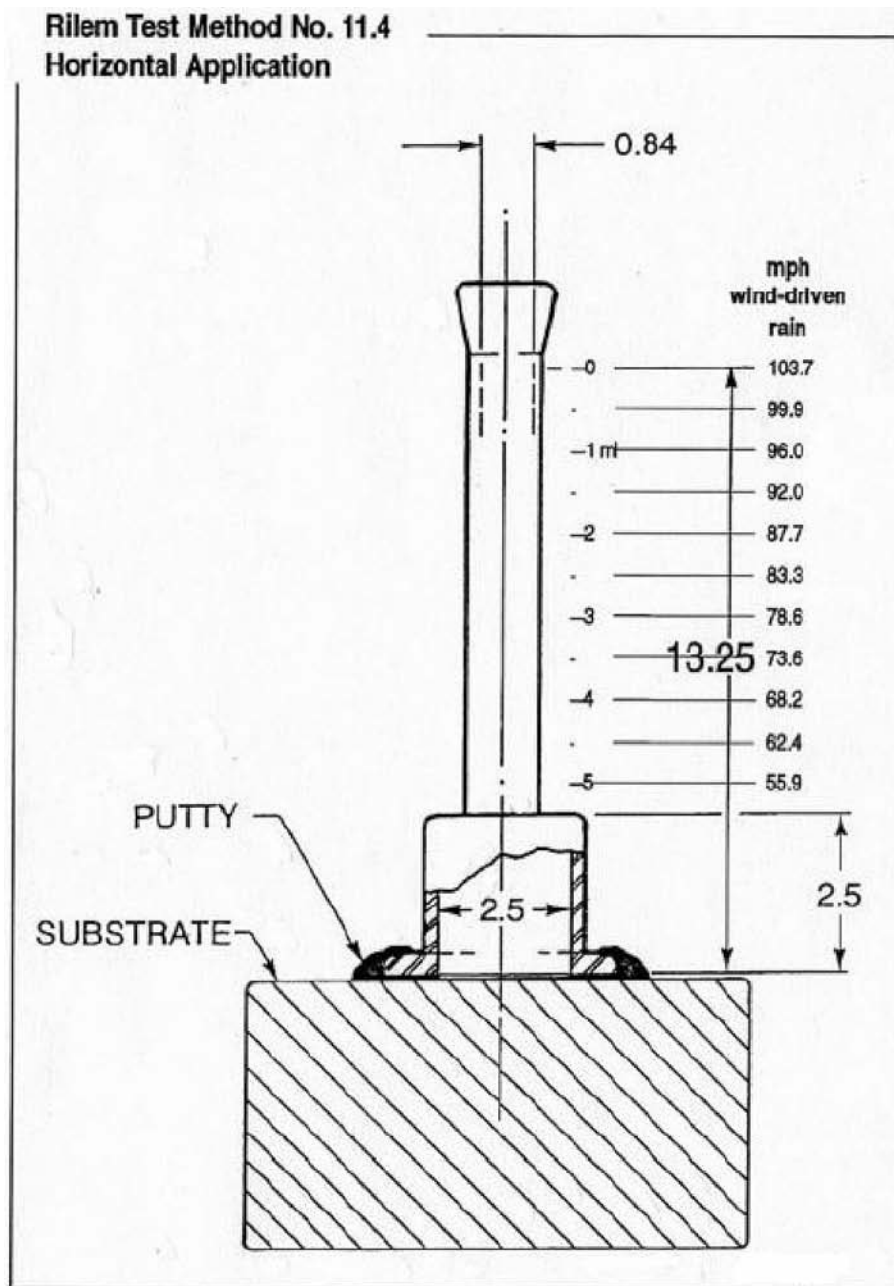


Figure 2. Experimental setup for application of Rilem Test Method^[14].

Figure 2. Diseño experimental para la aplicación del método de ensayo Rilem^[14].

2.2.4. Durability: Aging test

The salt mist test is an accelerated aging test that can cause a decline in mechanical strength and other physical and chemical properties. This test was performed according to standard UNE-EN 14147:2003^[16]. Tests were carried out in an accelerated salt spray chamber (ASCOTT Mod. 450T), where the samples were exposed to NaCl

solution and drying air at a constant temperature of 35 ± 5 °C. The test consisted of 60 salt mist exposure cycles, each comprised of 4 h saline water spraying and 8 h drying. After each 15-cycle period the macroscopic appearance of the samples was inspected to check for any kind of deformation. At the end of the test the samples were submerged in deionized water until the salt was fully removed (21 days, renewing the water each day) and then dried at

70 ± 5 °C until reaching a constant weight (11 days) before finally determining capillary absorption and mechanical strength.

2.2.5. Durability: Freeze-Thaw cycles

Resistance to freeze-thaw cycles was studied by the application of standard ISO 20394:2007^[17]. The method is intended to simulate freeze-thaw effects on insulating material which are frequently exposed to water and low-temperature conditions. The samples were subjected to 5 cycles consisting of 17 hours at -20 °C followed by 7 hours at room temperature, and subsequently to 30 cycles of a wet-dry process consisting of 12 hours dipped in water followed by 11 h in a muffle at 60 °C and 1 h at room temperature. Capillary water absorption and mechanical strength were determined at the end of the test.

2.2.6. Durability: Fire performance

Fire performance characteristics were studied by the application of standard EN ISO 11925-2:2002^[18]. The test was carried out on 40 x 40 x 160 mm monoliths (much smaller than the size indicated in the standard). Two samples were tested, Hg30SC and the reference sample. The samples were conditioned prior to testing for 48 h at 23 °C and 50 % humidity. The purpose of this test is to study how a product reacts when fire breaks out in a room (not when the fire is developing or when it reaches full power), i.e. it imitates the start of fire, when a limited area of the product is exposed to a low heat output flame (~ 1 kW) maintained for 30 s of exposure.

3. RESULTS AND DISCUSSION

3.1. Capillary water absorption

The principal results of the capillary water absorption test are shown in table I (column A), which reports values for capillary absorption coefficient (Ws) and mass variation (ΔM) after 28 days. The capillary absorption coefficient is very low for all the samples before any durability test, being lower for Hg-S concrete (20 % and 30 %) than for the reference. This finding may be attributable to the fact that metacinnabar can decrease the open porosity of the concrete by filling up the external pores. In Part I^[12], total porosity values of 7.67, 2.88 and 1.97 % were reported for the reference, Hg20SC and Hg30SC, respectively, this means that the Ws value decreases as the mercury content rises.

To evaluate the capillary water absorption rate in a Hg-S concrete (Hg20SC and Hg30SC) as a function of time it is necessary plot the variation in Ws versus the square root of time (Fig. 3). According to the slope of curves, three zones can clearly be seen in both Hg-S concretes. At first (zone A) both samples have a similar Ws coefficient (0.08 and 0.07) and a similar water absorption rate. In this zone it is possible to see a fast increase in capillary absorption from 10 min and 3 h, but it is important to note that the Hg20SC sample needs an induction period of about 2 h for the capillary absorption process to start. After 3 h the Hg30SC sample has a Ws coefficient of 0.7 g cm⁻². Between 3 h and 7 days (zone B) capillary absorption is constant for both samples, and the Hg30SC sample has a higher Ws coefficient than Hg20SC. Finally, in zone C, corresponding to the period between 7 and 28 days, a fresh increase in capillary absorption takes place, but

Table I. Results of Capillary Absorption Coefficient according UNE-EN 480-5:2006

Tabla I. Resultados del Coeficiente de Absorción por Capilaridad según UNE-EN 480-5:2006

Sample	Capillarity Absorption, Ws (g.cm ⁻²)		
	(A) Initial (as-obtained) samples	(B) After salt mist aging test	(C) After freeze-thaw and wet-dry test
SC	0.12	0.18	0.32
Hg20SC	0.08	0.04	0.11
Hg30SC	0.07	0.01	0.03

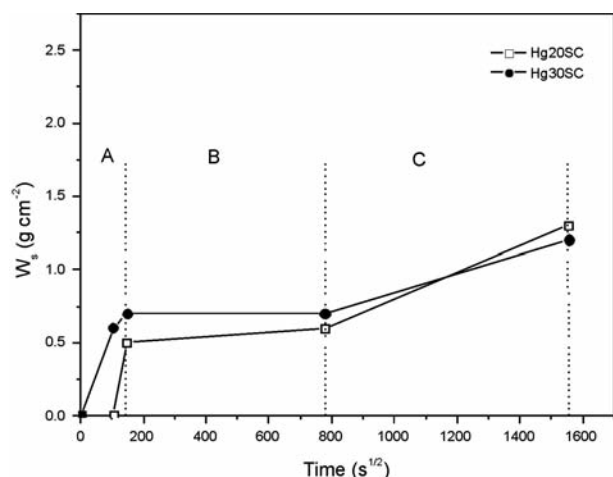


Figure 3. Variation in W_s versus $t^{1/2}$ for Hg20SC and Hg30SC.

Figure 3. Variación de W_s frente a $t^{1/2}$ para las muestras Hg20SC y Hg30SC.

in this period the slope is smaller than in the initial time period (zone A) and the W_s coefficient value is inverse of the zone A and B. At 28 days the Hg20SC sample has higher W_s than Hg30SC. This effect could be related with the different evolution of porosity in these materials over time, where Hg20SC has a higher open porosity than Hg30SC: 1.44 % vs 0.57 %, respectively. These results may be related to the density values measured in these samples^[12], where Hg20SC has a bulk density of 2.834 g cm⁻³ compared to 3.118 g cm⁻³ in the case of Hg30SC. This property is defined as the mass of particles of the material divided by the total volume they occupy. Probably as a result of the evolution of the Hg30SC microstructure, this sample has developed a denser matrix that affords it good durability in contact with water.

By analyzing zones A+B, i.e. up to 7 days, the curves fit well a kinetic model represented by equation (3).

$$W_s = C (t^{1/2}) \quad (3)$$

where C is the coefficient of capillarity and t the time. The C values calculated for Hg20SC and Hg30SC samples were 0.01 and 0.51 g cm² s⁻², respectively. Although the water absorption rate prior to the stabilization zone is higher for Hg30SC, the results for both samples are extremely low, and thus it may be considered that these samples do not absorb water by capillarity.

It is noted that this test is used for no longer than 3 days for most building materials. The capillarity value of an impermeable mortar ranges between 2-3 g cm⁻², which is very much higher than those obtained for the stabilized mercury samples obtained

with the SPSS process. In view these results, it may be considered that both Hg20SC and Hg30SC present good capillary water absorption behavior.

3.2. Water permeability

Water permeability is a property that is directly related to the degree of water penetration or water diffusion through the pore system. Capillarity is activated by the surface tension when water enters a capillary. Low pressure water permeability was tested from 2 to 180 min. The result was negative for all the samples, i.e. none of the Hg-S concrete samples absorbed water by low pressure. Traditional mortar cements present penetration values of between 4-8 mL, which is considered very high permeability. For traditional concrete the values range between 0.4 -1.0, which indicates low permeability^[19]. It is estimated that values below 0.1 mL (for 10 min) indicate that the material does not exhibit capillary activity. In view of these observations, it may be stated that both Hg20SC and Hg30SC are, from this point of view, good materials to immobilize mercury from the environment and ensure its stabilization.

3.3. Resistance to different pH values

The behavior of the Hg-S concrete samples in different pH solutions has been studied following the method described in standard UNE-EN 13580:2003^[15] as above described. Table II presents the results for the Hg30SC sample, which is that with the highest mercury content.

Table II. Results of Absorption Coefficient AR (%) of Hg30SC for different pH values according to UNE-EN 13580:2003^[15]

Tabla II. Coeficiente de absorción AR (%) de la muestra Hg20SC a diferentes valores de pH, según UNE-EN 13580:2003^[15]

pH value	AR (%)
2	0.22
4	0.35
6	0.34
8	0.34
10	0.40
13*	0.17

* This pH corresponds to the KOH solution according to UNE-EN 13580^[15].

The water absorption coefficient value is very low in all cases, ranging between 0.2 - 0.4 %. This means that the sample has very high resistance to both acid and alkaline aggressive environments.

Macroscopic observation of the sample after 42 days immersed in the different pH value solutions confirmed the absence of damage or deterioration on the surface. There was also no significant mass loss or surface roughness.

3.4. Aging test: Salt mist

Figure 4 shows the aspect of the samples after 30 days in the salt spray chamber. Salt crystallization was only

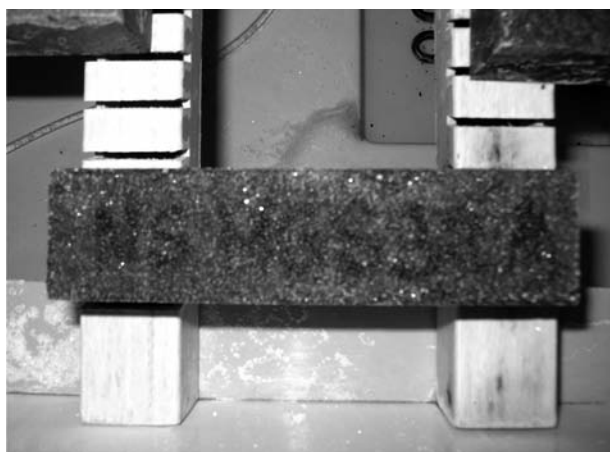


Figure 4. Aspect of Hg30SC sample after salt mist test.

Figure 4. Aspecto de la muestra Hg30SC después del ensayo en la cámara de niebla salina.

observed on the sample surface and did not give rise to cracks, surface scales or any kind of disaggregation of the materials.

After washing and drying as explained in section 2 above, the samples were subjected to a capillary absorption test, and the results are shown in table I (column B). W_s values indicate that the salt mist test did not increase the porosity of either the Hg20SC or Hg30SC samples.

With regard to compressive strength, table III (column B) displays the test results along with the initial values reported in Part I^[12]. The loss of compressive strength after highly aggressive salt mist aging (30 days in salt mist exposure at 35 °C, 21 days submerged in water for desalination and 11 days at 70 °C for drying) is negligible for Hg-S concrete (nil for sample Hg30SC and very low (5 %) for sample Hg20SC) but not for the reference sample, which exhibited a 22 % decrease in compressive strength.

The much better performance of the Hg-S concretes than the reference sample may be related to the lower porosity of the samples with mercury, as previously noted in Part I^[12].

3.5. Durability: Freeze-Thaw cycles

After 5 freeze-thaw cycles and 30 wet-dry cycles the samples presented no disaggregation of the material or cracks or scales. Variations in capillary absorption were quite negligible for Hg-S concrete, but a very slight increase was detected in reference sample, as shown in table I (column C).

Mechanical strength values fell by about 22 % for the Hg-S concrete and by nearly 40 % for the

Table III. Comparison between compressive strength of samples after durability tests.

Tabla III. Comparación de los valores de resistencia a compresión de las muestras después de los ensayos de durabilidad.

Sample	Compressive strength (N mm ⁻²)		
	(A) Initial (as-obtained) samples*	(B) After salt mist aging test	(C) After freeze-thaw and wet-dry test
SC	58.0 ± 2.7	45.2 ± 4.1	35.1 ± 1.4
Hg20SC	58.4 ± 4.2	55.4 ± 4.5	46.5 ± 2.9
Hg30SC	57.2 ± 4.4	57.2 ± 5.6	44.5 ± 6.9

* Data from^[12].

reference sample (Table III (column C)). As in the other tests the Hg-S concretes exhibited better resistance than the reference sample, even in this highly aggressive test.

3.6. Durability: Fire performance

The results of the fire test, table IV, show that a single-flame source of low heat output (~ 1 kW) maintained for 30 s of exposure did not give rise to the origination and spreading of any flame on the surface of either the Hg-S concrete or reference sample test specimens. Light clear-grey fumes were occasionally observed during the test and the surface exposed to the flame was slightly carbonized. This indicates that, in the experimental conditions of the standard test, the fire hazard of the samples is very low.

Table IV. Results of Fire Test for 30 s exposure to flame

Tabla IV. Resultados del ensayo de fuego para 30 s de exposición de llama

Parameter	Sample	
	SC	Hg30SC
Ignition of specimens (yes/no)	no	no
Time of ignition (s)	—	—
Vertical flame spread within 150 mm	no	no
Ignition of filter paper (yes/no)	no	no

4. CONCLUSIONS

- The materials obtained in the immobilization of mercury with SPSS technology present a concrete-like characteristic which confers excellent mechanical strength. Their low porosity affords very high impermeability, impeding any reaction with water or aqueous solutions, thus achieving high resistance in both acid and alkaline media.
- As the salt crystallization test and freeze-thaw plus wet-dry tests did not produce any deterioration, disaggregation of materials or loss of mass on the surface of the samples, this means that their

durability in a highly aggressive environment such as that tested in this work is very high.

- Although the test methods applied in this work were defined for other kinds of materials (building materials), due to the lack of a standard process to determine the long-term stability of immobilized waste, they may be considered contribute to that those can be a very useful tool to determine the durability of materials obtained by waste stabilization/solidification processes.

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REFERENCES

- [1] UNEP, Draft technical guidelines on the environmentally sound management of mercury wastes, 4th Draft, april, 2009. <http://www.basel.int/techmatters/mercury/guidelines/040409.doc>
- [2] Directive 2000/60/EC of the European Parliament and of the Council of 23 October 2000, Establishing a framework for Community action in the field of water policy. Official Journal L 327, 22/12/2000; 0001-0073.
- [3] Regulation (EC) No 1102/2008 of the European Parliament and of the Council of 22 October 2008 on the banning of exports of metallic mercury and certain mercury compounds and mixtures and the safe storage of metallic mercury, Official Journal of the European Union. L 304 (14/11/2008), pp. 75-79.
- [4] European Commission, Brussels, Requirements for facilities and acceptance criteria for the disposal of metallic mercury. Bipro. Final Report. 16, april, 2010.
- [5] R. Malviya and R. Chaudhary, *J. Hazard. Mater.* B137 (2006) 267-276.
- [6] J.M. Zhuang, T. Lo, T. Walsh and T. Lam. *J. Hazard. Mater.* B113 (2004) 157-164.
- [7] X.Y. Zhang, Q.C. Wang, S.Q. Zhang, X.J. Sun and Z.S. Shang, *J. Hazard. Mater.* 168 (2009) 1575-1589.
- [8] P. Randall P and S. Chattopadhyay, *J. Hazard. Mater.* B114 (2004) 211-223.
- [9] M. Fuhrmann, D. Relamed, P.D. Kalb, J.W. Adams and L.W. Milina, *Waste Manage.* 22 (2002) 327-333.

- [10] E.A. López, A. López-Delgado, F.J. Alguacil and M. Alonso, Spanish Patent P200930672, 2009.
- [11] F. Sandrolini, S. Manzim and A. Andrucci, *Compos. Part A-Appl. S.* 37 (2006) 695-702.
- [12] A. López-Delgado, F.A. López, F.J. Alguacil, I. Padilla and A. Guerrero, *Rev. Metal. Madrid* 48 (2011) 45-57.
- [13] UNE-EN 480-5: 2006 Admixtures for concrete, mortar and grout. Test methods. Part 5: Determination of capillary absorption. AENOR.
- [14] RILEM, Water absorption under low pressure. Pipe method. Test No. II.4, tentative recommendations. Paris, 1980.
- [15] UNE-EN 13580:2003 Products and systems for the protection and repair of concrete structures. Test methods. Water absorption and resistance to alkali for hydrophobic impregnations.
- [16] UNE-EN 14147:2003 Natural stone test methods, Determination of resistance to aging by salt mist.
- [17] ISO 20394:2007 Thermal-insulating materials. Determination of freeze-thaw resistance.
- [18] EN ISO 11925-2:2002 Reaction to fire tests. Ignitability of building products subjected to direct impingement of flame - Part 2: single-flame source test.
- [19] F.J. Alejandro, *Historia, caracterización y restauración de morteros*. Ed. Univ. Sevilla. Insto. Universitario de Ciencias de la Construcción. Sevilla, España, 2002, pp. 67-70.