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ORIGINAL PAPER

Stabilization/Solidification of Hazardous Metals from Solid Wastes into Ceramics

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Abstract Safe management of hazardous metals deriving from solid wastes constitutes an important environmental challenge. In the present study, recent progress in the stabilization/solidification of heavy metals from various solid wastes into ceramics and glass–ceramic materials is reviewed. Coal/lignite-fired power plant fly and bottom ashes, steel industry by-products, metallurgical and urban/municipal wastewater sludge, and pharmaceutical wastes are included. The environmental behavior of the ceramics produced is studied via several leaching tests and ecotoxicological analyses. The goal is to assess the potential of these materials for retention of pollutants into their microstructure, for safe use as building materials. These objectives are in line with green chemistry and sustainable development principles. Moreover, the aim of turning waste into useful feedstock for another industrial sector towards symbiosis, ample coordination, and circular economy, is strongly encouraged by current environmental policies. Such secondary resources can be considered as substitute materials in ceramics production, as they contain useful oxides. However, the presence of heavy metals remains a significant environmental issue. Possible leaching of pollutants caused by rain from construction materials incorporating solid wastes would lead to contamination of surface and subsurface water, thus raising critical environmental concerns. Inertization of potentially hazardous metals into environmentally-friendly ceramics and glass–ceramics is shown to be feasible, as concentrations of these elements in the leachates remain within acceptable limits. Certainly, this also depends on the leaching method employed and system parameters. In conclusion, stabilization of these elements into ceramic matrices can be an effective and beneficial alternative for protecting human health and the environment.

Keywords Stabilization/solidification · Solid waste · Sludge · Hazardous metals · Leachability · Ceramics

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Introduction

The stabilization/solidification (S/S) of industrial wastes into clayey raw materials used in standard ceramic manufacturing can contribute to environmental protection and sustainable development. Actually, special emphasis is placed on resources optimization, in order to (a) minimize uncontrolled waste disposal that represents a significant potential source of release of environmentally sensitive elements to the environment [1], and (b) reduce exploitation of natural raw materials, towards eco-friendly building materials production. Moreover, value-added ceramic and glass–ceramic products can be obtained, which satisfy quality and technical requirements for a market of increasing competitiveness.

Industrial solid by-products can, actually, be considered as substitute materials in ceramics fabrication due to their valuable oxides content. However, the presence of heavy metals, including As, Hg, Cd, Co Cr, Br, Sr, Pb, Se, Sn and Zn, constitutes a significant environmental issue. So far, the environmental compatibility of several industrial secondary resources has been evaluated. Fly and bottom ashes produced in huge quantities in thermal power plants fed with high ash content low grade coal/lignite, and also steel industry electric arc furnace dust, various sludges from different origins, etc. have been considered. Waste stabilization is needed to avoid serious environmental concerns upon landfilling.

For this purpose, various standard leaching tests, including EN12457-2 (European), TCLP and ASTMD3987 (American), DIN38414-S4 (German), NEN7341 (Dutch), AFNORX31-210 (French), NBR10005 (Brazilian), and JST-13 (Japanese) have been employed. Certainly, the leachability of the compounds depends on the leaching test used. Moreover, ecotoxicological characteristics of some industrial solid wastes have been determined using bioassays [2–6]. Because of the mobility and bioavailability of many of the aforementioned elements upon water extraction, possible leaching of pollutants caused by rain can lead to contamination of surface and subsurface water, which represents a critical environmental concern. The mobility of most elements contained in combustion ashes is dominated by pH, which, in turn, is strongly influenced by the amount of calcium present. Particularly in fly ash, calcium cations can exhibit the highest extraction ratio, regardless of the leaching test employed [7–12]. Furthermore, for the determination and better understanding of the mineralogical and morphological transformations that may occur during the leaching process, not only the leaching eluates but also the solid residues of the leaching test should be analyzed. In fact, phases such as ettringite and/or calcium silicate hydrates are reported to precipitate in the solid residues of some fly ashes after leaching tests, which can be environmentally beneficial, as ettringite immobilizes several toxic elements including As, Cr, Se, and Sb into its structure [13].

Leachability of pharmaceutical wastes (pharmaceutical industrial sludges) was also studied and the need for the implementation of S/S processes for their safe utilization into environmentally friendly value-added materials was highlighted [14]. Besides, the leaching of hazardous metals from specific wastes such as expired pharmaceutical wastes and their ashes should also be studied, in order to assess the environmental impact after their disposal by direct landfilling or incineration techniques. Recently conducted study showed that Cd, Ni, and Pb exceeded the pertinent limits (Decision 2003/33/EC) in ashed drugs leachates (EN14429), while raw expired drugs as well as raw and ashed packaging did not exceed the corresponding limit values, and, therefore, could be disposed of directly to a non-hazardous waste landfill. In all cases, the highest leaching potential was measured at acidic pH (<4) [15]. Furthermore, medical waste generated by health-care activities comprise not only infectious, pathological, chemical, pharmaceutical, genotoxic and radioactive wastes but also heavy metals (World Health Organization), and have attracted a lot of research attention during the past decades [16].

From the environmental point of view, the physicochemical processes, the interrelationships, and the assessment of the accumulation of contaminants including heavy metals into soil and subsurface have been extensively studied for predicting the fate of the pollutants and the risk they pose to human and ecological receptors, so far [17–19].

In the present study, recent progress in the S/S of potentially hazardous heavy metals from various solid wastes into fired ceramic materials is reviewed. Possible immobilization of these potentially toxic elements into ceramic matrices encapsulating industrial powdery residues, will avoid leaching, water resources contamination and bioaccumulation of trace pollutants. Thus, it can be much beneficial for protecting human and environmental health. The environmental behavior of these ceramics upon subjection to several leaching tests and ecotoxicological analyses is studied. The potential for retention of such pollutants into clay-based ceramics towards their safe use as building products is assessed. These objectives are in line with, at least, two of the Twelve Green Chemistry Principles, particularly with the 3rd paragraph ("Less Hazardous Chemical Syntheses") and the 7th one ("Use of Renewable Feedstocks") [20]. It should also be emphasized that the aim of turning waste from one industry into useful feedstock for another one is strongly encouraged by current E.U. environmental policies for sustainable development.

Stabilization/Solidification (S/S) of Hazardous Metals from Solid Wastes into Ceramic and Glass–Ceramic Microstructures

Several studies are reported in recent literature on the leaching behavior of heavy metals from different industrial solid waste by-products that have been stabilized/solidified in standard clayey raw materials used in the manufacture of resource efficient and lower cost ceramics or glass–ceramics. Their physico-mechanical and chemical properties are not significantly affected by the incorporation of the industrial by-products. The high rate of utilization of ceramics in the building sector enhances the importance of the endeavor. In most cases, the pH of the medium, varying with the leachant used according to the evaluation test employed, appears a determinant parameter for dissolution of metals, governing their release from the solid phase into solution. From the technological point of view, success in waste recycling frequently depends on controlling compositional variations [21], thus maintaining a purity level in the waste streams.

Industrial Solid Residues

In particular, environmental properties including the leaching properties followed by ecotoxicological analyses of ceramics sintered via conventional technology or novel two-step method and also microwave irradiation have been recently investigated. Extruded clayey mixtures encapsulating industrial powdery by-products, specifically *lignite* power station *fly* and *bottom ashes* derived from *conventional burners* and also *circulating fluidized bed combustion* (*CFB*) *ash* were utilized [22–25]. The goal was to assess the potential for safe use of the ceramics obtained as building products. The ceramics produced would not be characterized as ecotoxic, because the ecotoxic effect of the leachates of crushed materials did not exceed 20 %. Petrurgic method at 950 °C for a mixture of fly and bottom ash was also proposed as a simple, inexpensive and energy saving process for the development of engineering and construction applications, as confirmed by the concentration of heavy metals upon leaching tests [26]. Moreover, the amount of heavy metals released from macroporous ceramic pellets with controllable degree of pore connectivity prepared from fly ash via mechanical foaming and pseudo-double-emulsion method, was showed to be much lower than that from fly ash powders. This is due to heavy metals melting and subsequent immobilization as well as their partial sublimation during the sintering process [27]. Furthermore, vitrification was proved to be an effective method for a successful immobilization of heavy metals into the microstructure. In fact, glass–ceramics produced from thermal transformation of fly ashes derived from lignite and bituminous coal-fired boilers show amorphous texture and are characterized by low leachability [28]. Hazardous elements retention efficiency is also obtained by the valorization of high calcium lignite fly ash, as well as red mud, ferronickel slag and steel slag, in the development of glass–ceramics [29, 30].

Valorization into glass, glass–ceramics and ceramics represents a potential application related to the reuse of *fly ash* also from *municipal solid waste incinerators* (*MSWI*). A systematic comparative analysis of different alternative options shows new possibilities for utilization of MSWI ashes in this field, which can possibly lead to great advantages in waste minimization [31]. Certainly, it is important to prevent heavy metal-rich wastes from being incinerated for reducing the content of toxic metals in fly ash, as the heavy metal content of fly ash was determined to be a function of the solid wastes incinerated [32]. In order to evaluate the effectiveness of heavy metal immobilization into ceramic or glass–ceramic matrices, MSWI fly ash was even treated with microwave sintering towards glass–ceramics development, using different additives (Al_2O_3 powder, γ-Al_2O_3, SiO_2 and kaolin). Leaching tests in the glass–ceramics produced show that heavy metals are effectively immobilized especially in samples containing aluminosilicates as the main crystal phases [33]. Moreover, the solidification of hazardous metals from *MSW gasification slag* was studied in non-burnt bricks, where the heavy metals (As, Cd, Cr, Ni, and Zn) appear to be effectively inertized in form of oxidizable and residual fractions, stable and not easy to dissolve. On the basis of this finding, the risk of exposure to heavy metals and the impact on the ecological environment of the non-burnt bricks developed is evaluated to be relatively negligible, whereas attention is only warranted for potential migration of cadmium in the long term [34].

Besides, mixing *fly ash* from *clinic* and *hazardous* waste incineration and energy production plant with clayey raw materials, as admixture to brick production process up to 20 %, appears to be a convincingly suitable process, both for the stabilization process and the technological properties of the final product [35].

Electric arc furnace dust (*EAFD*), is generated upon steel scrap melting in the steelmaking industry via the volatilization of metal particles that are oxidized, solidified and detained in specially designed filters, in the form of fine powder. EAFD can be regarded as a material of no environmental concern, when it is stabilized in non-metallurgical industries, such as the structural clay ceramics industry. In fact, EN12457 standard leaching tests [36] of mixtures of clay with 2.5 and 5 wt% EAFD content, fired at 850, 900 and 950 °C, showed that the quantities of heavy metals leached from crushed blocks are within regulatory limits [37]. Other researchers also implied that using steelmaking dust in the manufacture of ceramic construction materials, such as clay bricks, ceramic ware and cement bricks, is both possible and reasonable [38, 39]. The mechanism of stabilization of heavy metals was also investigated using Toxicity Characteristic Leaching Procedure (TCLP) tests [40] in construction materials prepared in the form of bricks and aggregates through a conventional ceramic processing (extruding) method and sintered at 950–1050 °C for 3 h. For that purpose, a mixture of industrial dust wastes, namely *EAFD*, *fly ash*, *stone ash*, were introduced into clayey raw materials, allowing a compositional variation. Pretreatment processes including ion exchange, control of electrolyte concentration and pH are showed to be important for heavy metals stabilization in the raw material mixture [41]. Moreover, utilization of other by-products of the electric arc furnace steelmaking process, specifically of *electric arc furnace slag* (*EAFS*) and *ladle furnace slag* (*LFS*), as admixtures into clay-based ceramic construction products fired at various temperatures to be used for protection from electromagnetic environmental pollution, is possible, as leaching test performed showed stabilization of all studied toxic elements within the sintered ceramic structure [42]. Environmentally friendly incorporation up to 9 wt% of *mill scale*, another steel industry waste, produced during the milling process from the rapid oxidization of the hot iron products, as an efficient secondary resource into red ceramics is also feasible [43].

Various by-products of other metallurgical industrial activities, which are currently problematic wastes, are also under consideration for S/S into ceramic matrices. In particular, leaching tests led to the conclusion that 50 and even 70 wt% of *red mud*, an alkaline leaching waste from *bauxite* processing plant, can be properly used with heavy clay in the production of pressed ceramic bodies sintered at 900–1190 °C [44]. Also, improvements in potential leachability of some pollutants can be attained by the incorporation of *Waelz slag* and *foundry sand* into clay-based bricks fabricated in a semi-scale industrial trial. Additions to less than 30 wt% were proved to meet regulatory leaching limits [45]. Furthermore, by recycling metallurgical waste mixtures, including *foundry sands*, *exhaust metallurgical dust*, spent *galvanic glass microspheres* and *acid inertization salt*, into ceramic materials containing 75–85 % of the industrial wastes and fired at 950–1010 °C, heavy metals leaching and solubility levels remained hundreds of times less than those permitted by national standards. This is due to the formation of a glassy structure with inclusion of newly formed minerals [46]. Galvanic sludges are derived from physico-chemical treatments of electroplating plant wastewaters. The leachability of relatively large amounts of toxic metals found in industrial *sludges* generated in *galvanizing* processes can be reduced to negligible levels after utilization of the sludges as raw materials into fired ceramics for possible building applications [47–52]. The effective inertization process of some heavy metals of these sludges, such as Pb and Zn, should be attributed to their volatilization as far as the ceramic firing temperature is increased, followed by their encapsulation in aluminosilicate phases into the ceramic matrix during solidification [53, 54].

Various Sludges

A method for fabrication of glass–ceramics from *heavy metal gypsum* and *pickling sludge*, as well as for stabilization of Pb, Zn, Cd, As, Hg, Cr and Ni heavy metals, was also proposed. This leads to a toxic elements leaching lower than the TCLP method requirements. At this end, heavy metal gypsum was desulfurized and intermediate products formed were treated with the pickling sludge by the conventional heating method (nucleation at 700 °C for 2 h and crystallization at 900 °C for 1 h) for the preparation of glass–ceramics [55]. Solidification/stabilization of chromium and nickel contained in pickling sludge, one of stainless-steel making by-products by its reusing (14 wt%) as a nucleation agent of glass–ceramics was also examined, leading to crystallization temperature decrease by 110.8 °C, as well as microstructural and mechanical performance improvements, while the safety of the process was confirmed by TCLP testing results [56].

Textile industry wastewater treatment *sludge* is often contaminated with heavy metals of dyestuffs and chemicals, and, therefore, are considered hazardous. Mixing with clay can be an eco-friendly solution for the management of textile laundry wastewater sludge, incorporated up to a 20 wt% to produce safe and inert bricks for civil construction, according to the results of the heavy metals leaching and solubilization tests [57]. Incineration techniques can also be used for as much as 80 % textile sludge volume reduction and destruction of the hazardous elements. Then, S/S of up to 10–20 wt% of the incinerated ash into ceramic tiles and blocks can be achieved [58]. The detoxification of chrome sludge taken from an iron and steel plant by manufacturing bricks was also proposed, thus turning another industrial waste into resource and also making economical benefits. The optimum conditions for toxicity reduction by transforming hexavalent chromium into trivalent chromium as well as for the production of good quality bricks were to mold (20 MPa) and sinter (950 °C, 4 h) a chromium sludge, coal and clay mixture at the ratio of 8:82:10 % [59].

Furthermore, *petroleum sludges*, waste materials discarded in large amounts by the petroleum exploration industry, have also been proposed for incorporation into clay bodies up to 30 wt% for clay-based ceramic pieces manufacturing (700–1100 °C). They can also replace natural kaolin material up to an optimum of 2.5 wt% for high-quality porcelain stoneware tile production by pressing and then firing at (1240 °C). The leaching concentrations of Ag, As, Ba, Cd, Cr, Hg, and Pb of the sintered ceramics are proved to meet the national regulatory requirements. This offers a new possibility for an environmentally-friendly management of this abundant solid waste, thus potentially contributing to alleviating the environmental impact of this industrial sector [60, 61]. On the other side, *spent fluid catalytic cracking catalysts*, hazardous solid wastes generated by *petroleum refineries*, and containing vanadium and nickel, were efficiently mixed with marine clays to produce fired bricks with low metal leachability. This is due to stabilization and encapsulation mechanisms involving formation of physical barriers around the heavy metals or even bonds between the marine clay matrix and the metals in the ceramic microstructure [62].

Table 1 Stabilization/solidification (S/S) of hazardous metals from solid wastes into ceramics and glass–ceramics

Wastes	Pollutants	Methods	Conditions	Products	Leaching test	References	Comments
Fly and bottom ashes derived from conventional burners and also CFB-combustion ash	Heavy metals	Sintering via conventional technology or microwave irradiation	Extruded up to 20 wt% fly ash Compacts: 100 % 850–1150 °C	Building ceramics	European standard test EN12457-2:2002	Karayannis et al. [22, 24] Koukouzas et al. [25]	Compacted: 100 % Extruded clayey mixtures encapsulating industrial powdery by-products
Mixture of fly and bottom ashes	Heavy metals	Petrurgic method	950 °C	Possible engineering and construction applications	TCLP tests (EPA1992) pH 4.93 ± 0.05	Vu et al. [26] Banar et al. [35]	Glass–ceramic form: 80 % bottom ash and 20 % fly ash
Industrial solid residues	Heavy metals	Slurry preparation, foaming, pelletizing, drying, and sintering	600–1200 °C	Macroporous ceramic pellets	TCLP tests pH 4.93 ± 0.05	Han et al. [27]	Average size of macroporous flyash pellets
Fly ashes from thermal power plants fired by lignite and bituminous coal	Heavy metals	Vitrification	Heat treatment	Glass/glass–ceramics	European standard test EN12457-2:2002	Kasprzyk et al. [28] Karamberi et al. [29] Kritikaki et al. [30]	Amorphous texture/low leachability caused by successful immobilization of the heavy metals into the glass structure
Municipal solid waste (MSW) fly ashes	Heavy metals, toxic organic substances (dioxins, furans and PAHs)	MSW gasification slag non-burnt bricks, chemical immobilization	1540–1600 °C 1300 °C	Construction materials: (cement, concrete, ceramics, glass and glass–ceramics), geotechnical applications (road pavement, embankments), “agriculture” (soil amendment); and, miscellaneous (sorbent, sludge conditioning)	EPTOX toxicity procedure/TCLP China TLCP (China leachability toxicity standard method) Four-step EBCR sequential extraction procedure)	Ferreira et al. [31] Youcai et al. [32] Chou et al. [33] Wang et al. [34]	Framework of best technologies and final products, waste minimization and resources conservation
Electric arc furnace dust (EAFD), fine powder, generated upon steel scrap melting for steelmaking	Zn, Pb and Cd/hazardous high metal content waste	Volatilization of metals that are oxidized and subsequently solidified	2.5–5 wt% of EAFD 850–950 °C	Ceramics/blocks	European standard test EN12457-2:2002 pH 6.2–12.6	Stathopoulos et al. [37]	Stabilization properties of EAFD/clay ceramic structures and the potential of EAFD utilization into structural ceramics production (blocks)

Table 1 continued

Wastes	Pollutants	Methods	Conditions	Products	Leaching test	References	Comments
Fly ash samples	Heavy metals	Sintering via conventional technology	850–1050 °C	Ceramic construction materials: clay bricks, ceramic ware and cement bricks	European standard test EN12457-2:2002	Karayannis et al. [38] Lis and Nowacki [39]	Up to 4 % of Zn and more than 20 % of Zn
Metallurgical waste mixtures (exhaust metallurgical dust, spent foundry sands, galvanic glass microspheres, and acid inertization salt)	Heavy metals	Sintering process	75–85 wt% of industrial wastes 950–1010 °C	Ceramic materials/bricks	Leaching and solubility with demands of the Brazilian standards	Kim et al. [41] Mymrin et al. [46]	All industrial wastes under study have high contents of heavy metals, such as Pb, Br, Sr and Cr
By-products of the electric arc furnace steelmaking process electric arc furnace slag, ladle furnace slag, pickling sludge	Heavy metals (Cu and Ni)	Landfill and cement solidification	850–1050 °C	As admixtures into clay-based ceramic, construction products, glass–ceramics		Bantsis et al. [42]	Protection from electromagnetic environmental pollution
Mill scale (MS)	Iron oxides	Extrusion and sintering processes	Clay/MS mixtures: up to 9 wt% of MS 1100 °C	It is also feasible as an efficient secondary resource into red ceramics	European standard test EN12457-2:2002	Spiliotis et al. [43]	Produced during the milling process from rapid oxidization of the hot iron products; steel industry waste, iron oxides >95 %
Red mud, from bauxite processing plant	Environmental impacts caused by the aluminum industry	Pressing with clay	50–70 wt% of red mud 900–1190 °C	Heavy clay ceramic bodies	Technical standard NBR 10005	Hildebrando et al. [44]	Alkaline leaching waste: SiO_2 (51.8 %), Al_2O_3 (24.5 %) and Fe_2O_3 (10.9 %) and ($Na_2O + K_2O < 0.35$ %)
Waelz slag and foundry sand	Heavy metals, Ba, Cr and Pb, reduced leaching of Ba and Pb	Incorporation into clay-based bricks Extrusion and sintering	<30 wt% of waelz slag	Red clay-based bricks	NEN 7345 diffusion leaching test equilibrium leaching EN 12457-1 and 2	Quijorna et al. [45]	Fabricated in a semi-scale industrial trial

Table 1 continued

Wastes	Pollutants	Methods	Conditions	Products	Leaching test	References	Comments
Galvanic sludge wastes (GSW)	Heavy metals	Extruding the mixture	980–1050 °C	Clay bricks, glasses	Method 1311-1 TCLP 1992 Product Consistency Test (PCT) or ASTM C-1285-94	Pérez-Villarejo et al. [47] Perez et al. [48] Silva and Mello-Castanho [49]	GSW in the clay–GSW bricks was up to 5 wt%
Sewage sludge from wastewater treatment	Oxyanion forming heavy metals As, Cr, Mo, Sb, Se, V and W	(Co)precipitation, adsorption, ion exchange and membrane techniques	1100 °C Heat treatment	Ceramic materials	EN 12457	Verbinnen et al. [50]	Sludge-adsorbent mixture
Sludge from etching and electroplating plants	Heavy metals (Cu and N)	Sintering	500–950 °C	Bricks	TCLP test	Tsai et al. [51]	
Industrial wastes	Heavy metals	Petrology	1250–1560 °C	Ceramics		Moroni et al. [52]	Inertization of highly chromiferous and Cr–Pb-rich industrial wastes
Industrial sludge/masonry clay/organic and inorganic industrial waste	Relatively large amounts of toxic elements	A hot-dip galvanizing process after wastewater neutralization	Thermal treatment 850–1000 °C	Bricks	SRPS EN 12457: compliance test for leaching of granular waste materials and sludges	Arsenovic et al. [53, 54]	3 and 6 wt% of sludge with 97 and 94 wt% of usual masonry clay
Heavy metal gypsum and pickling sludge	Heavy metals	Desulfurization of heavy metal gypsum and conventional heating method	700 °C (nucleation) 900 °C (crystallization)	Glass–ceramics	Environmental Protection Agency (EPA) 1311 method TCLP test regulatory standards	Pan et al. [55] Yang et al. [56]	The main crystalline phase of the obtained glass–ceramics is akermanite.

Table 1 continued

Wastes	Pollutants	Methods	Conditions	Products	Leaching test	References	Comments
Textile laundry wastewater sludges	Heavy metals		Up to 20 wt% of sludges (mass basis)	Safe and inert bricks for civil construction	Brazilian standards NBR 10005/04 and NBR 10006/04	Herek et al. [57]	Wastewater sludge until a concentration of 20 % (mass basis)
Ash after textile sludge volume reduction	Heavy metals of dyestuffs and chemicals	Stabilization/solidification by incineration techniques	500, 700 °C and finally at 800 °C 10–20 wt% fly ash	Ceramic tiles and blocks	TCLP test	Iqbal et al. [58]	Various proportions (0, 10, 20, and 30 %) with clay
Chromium sludge	Cr	Sintered via conventional technology	850–1050 °C	Ceramic construction materials: clay bricks	National standards (HJ/T 301-2007)	Yang et al. [59]	Coal and clay were used at the ratio of 0–12:90–78:10 %
Solid petroleum waste (SPW)	Ag, As, Ba, Cd, Cr (total), Hg, and Pb/V and Ni	Pressed and fired by using a fast-firing cycle	1240, 700–1100 °C	Porcelain stoneware tile body, clay ceramic pieces	NBR 10004 Brazilian standards pH 5.0 (acetic acid buffer)	Souza et al. [60] Pinheiro et al. [61] Sun et al. [62]	Replacement to natural kaolin material by up to 5 wt%, stabilize vanadium and nickel
Pet coke (PC) and sewage sludge (SS)		Extrusion and sintering		Clay-based ceramics	European standard test EN12457-2:2002	Spiliotis et al. [63]	Energy savings and improved thermal insulating properties
Sewage sludge from waste water treatment plants (WWTP)	Heavy metals	Extrusion and sintering process	980–1050 °C	Brick	National standards Neutral and acidic conditions Norm NEN 7345 from the Netherlands Tank Leaching Test (1993)	Zhang et al. [64] Cousido et al. [65]	Variable percentages of clays, sludges, forest waste (sawdust)

Table 1 continued

Wastes	Pollutants	Methods	Conditions	Products	Leaching test	References	Comments
Arsenic–iron sludge waste	Arsenic, iron	Extrusion process	1000 °C	Clay bricks	TCLP pH 3.0–12.0	Hassan et al. [66]	Optimum clay–sludge bricks was found to be 6 %
Dredging spoils	Heavy metal ions and organic pollutants	Sintering and the densification process	1200–1350 °C	Inert glass, glass ceramic products	National standards pH 5.0 (acetic acid buffer)	Bernstein et al. [67]	Mixture of calcinated sediment (80 wt%) and glass cullet powder (20 wt%)
Aggregates from construction and demolition waste (CDW)	Chromium and sulphate	Bricks, tiles, bluebirds and stoneware		Construction materials	TCLP test	Rey et al. [68]	Landfill Directive criteria

TCLP Toxicity Characteristic Leaching Procedure

The aforementioned industrial sludge wastes have a significant potential for inertization in the production of clay ceramics. Also, stabilization of heavy metals contained in *urban/municipal sewage treatment plant sludges* by solidification during the ceramic sintering process has attracted much attention. It is expected to provide an alternative to sludge discharge into landfill, despite some possible prejudices on the part of users. Actually, the water treatment plant sludge can be a potential substitute for clayey raw materials in ceramic manufacturing, as it is close to clay chemical composition [63]. In fact, the solidification for Cr, Cu, Zn, As, and Pb into sludge bricks obtained by mixing sewage sludge with shale was showed to be feasible in recent research. The leachate toxicity was proved to be lower than that of national standards, while a certain variation in leaching behavior is observed under neutral, acidic, or alkaline conditions [64]. The environmental compatibility of ceramics incorporated with wastewater sludge not only during their future life cycle as building materials but also after prospective deconstruction of the building after its useful life end, was confirmed by leachability and toxicity tests reported by other researchers [65]. Furthermore, fired bricks are made using arsenic–iron sludge generated from wastewater treatment systems as admixture to avoid accumulative rise of As and Fe concentrations by uncontrolled sludge dumping. Their leaching characteristics determined with the variation of pH reveal an environmentally-safe maximum amount of sludge that could be mixed with clay to produce clay-sludge bricks of acceptable technological properties [66]. Production of ceramics by vitrification at 1200–1350 °C was even proposed for the successful inertization of dredging spoils, material from *sediment* excavated from lagoon, containing several heavy metals and organic pollutants, thus being classified as toxic waste. For adjusting the compositional variations of the different sediment batches, *glass cullet* originating from a community glass recycling program, was used as admixture. The economic valorization of the waste was further enlarged by subsequent processing of the glass derived from vitrification by viscous phase sintering for crystallization and development of sintered glass–ceramic products suitable for building applications [67].

Specific Waste

Environmental assessment of specific waste such as *construction recycled materials* according to the Landfill Directive criteria reveals chromium and sulphate to be the most critical pollutant constituents of leachates, mainly attributed to be released by bricks and tiles rather than by recycled aggregates. However, ceramic materials were found to mainly release total Cr at the oxidation state of

Cr(III), whereas recycled aggregates mainly appear to release Cr(VI) that is extremely toxic. This highlights the need for distinguishing the oxidative state of chromium leaching level regulations in environmental legislation [68].

Furthermore, with regard to final product quality, presence of water soluble salts into ceramic materials incorporating additives, such as ammonium lignosulphonate, barium carbonate and calcarenite, can cause efflorescence phenomena that are among the most frequent problems, especially on face bricks, affecting their aesthetic quality [69–71].

Table 1 provides a critical summary of studies that produced different ceramics for the S/S of various solid waste materials, using various methods and conditions.

Concluding Remarks

Stabilization/solidification of potentially hazardous heavy metals of solid wastes including power station ashes, steelmaking by-products and various types of metallurgical wastes and sewage sludges into clay-based ceramics is feasible. The results reported in several relevant studies show an affinity between clay and solid wastes containing useful oxides, leading to optimized manufacturing of ceramics or even glass–ceramics with relatively low lixiviation levels, as concentrations of these elements in the leachates remain within acceptable limits, thus providing an alternative to minimize the environmental impacts caused by several industrial and urban activities.

Solubility appears to be an important factor controlling the leaching behavior. The potential of heavy metals to be transferred to the liquid phase primarily depends upon:

- the leaching method used and
- the pH of the medium, which varies with the leachant used and appears to be a determinant parameter for the dissolution of the hazardous elements.

Immobilization of various volatile (at firing temperature) elements of wastes is facilitated by presence of silica and alumina in the clays, which act as efficient "solvents" in the ceramic matrix.

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