Recycling difficult-to-treat e-waste cathode-ray-tube glass as construction and building materials: A critical review

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ABSTRACT: Cathode ray tubes (CRTs) waste generation has become a great environmental challenge worldwide. CRT glass possesses reasonable intrinsic strength, low water absorption and rich in silica, which makes the glass suitable for use as sand or pozzolan in construction materials. This work presents a comprehensive overview of literature reporting on the reuse of CRT glass to prepare glass-ceramics; cement mortar, paste, and concrete; and bricks. The effects of various critical factors on the resulting products' performance, preparation mechanisms, leaching behavior, lead fate, and environmental and human safety were investigated. The comparison of these recycling methods, and directions for future research were discussed and reported as well. Preparing cement mortar, paste, and concrete from CRT glass offer added advantages in terms of quantity of recyclable cathode ray tube glass at a given time, with minimal environmental and economic implications and thus could be an a promising value-added uses for CRT glass. The geographical distance between waste CRT glass sources and processing facilities, public policies should be taken into account in its recycling.

Keywords: E-waste; leaded glass; glass-ceramic; cement; lead recovery; leaching behavior

1. Introduction

The management and treatment of electronic waste (e-waste) has create a global environmental challenge, due to its rapidly growing volume and complex (or hazardous) nature. A report from "The Global E-waste Monitor 2014: Quantities, flows and resources" released by the United Nations University revealed that a total of an epic 41.8 million tons of e-waste was generated worldwide in 2014 (Baldé, 2015; Wang et al., 2016). In recent years, the replacement of cathode ray tube (CRT) sets with liquid crystal displays (LCDs), light-emitting diode (LED) panels and plasma display panels

(PDPs) is dramatically progressing, producing millions of units of waste CRTs. Data from the waste electrical and electronic equipment (WEEE) collection and pretreatment market indicate that approximately 50,000-150,000 million tons/year of end-of-life CRTs are currently collected in Europe, and this volume is not expected to decrease for coming several years (Andreola et al., 2007). In China, the recycling and dismantling amounts of waste electrical appliances (including TVs, refrigerators, washing machines, air conditioners, and personal computers) reached 41.499 million units in 2013, of which around 92% were TV CRTs. The bulk of a CRT consists of glass parts (including funnel, panel and neck glass), typically representing 85% of the total weight of monitors (**Fig. 1**). In the UK alone, more than 100,000 tons of CRT glass have been disposed of annually since 2003 (König et al., 2011). Globally, it is estimated that only about 26% of the discarded CRTs are recycled and the remaining 59% are landfilled due to less practical recycling approaches (Rashad, 2014). The panel made of barium-strontium glass, the funnel made of lead silicate glass containing approximately 20 wt% PbO and neck glass 40 wt% PbO (Yuan et al., 2012; Yu-Gong et al., 2016). Strong concerns have been raised about the potential of toxic-metal leaching from CRTs (Jang and Townsend, 2003; Spalvins et al., 2008). Therefore, there is a pressing need to develop effective recycling methods for these difficult-to-treat e-waste products.

Fig. 1 The generation of CRT glass

In general, there are two principal approaches of recycling CRT glass: closed-loop and open-loop recycling. In the closed-loop recycling, CRT glass is generally reused as raw material to manufacture new CRT monitors. For this recycling, it could be profitable only in the case of an absolute separation of the lead-containing and lead-free glass (Hreglich, 2001). With the rapid shrinking of demand for new CRTs, most CRT manufacturers have gradually ceased or restructured the funnel manufacturing facilities of their CRT operations. Therefore, a dramatic drop in closed-loop recycling has occurred, and attention has shifted to open-loop recycling (Mostaghel and Samuelsson, 2010; Mueller et al., 2012).

2. Construction and building materials

CRT glass possesses reasonable intrinsic strength, low water absorption and rich in silica, which makes the glass suitable for use as sand or pozzolan in construction materials. Accordingly, a number of projects have been undertaken to use CRT glass for the production of foam glass, ceramic-glaze, cement and concrete. There has been considerable research focuses on the feasibility of applying these recycled CRT wastes in the field of construction and building materials. However, to the best of our knowledge, there are sparse comprehensive reviews undertaken on this important topic except of the work of Rashad (2015), Iniaghe and Adie (2015). Hence this paper presents a thorough overview of the literatures reporting on the reuse of CRT glass to prepare glass-ceramics; cement mortar, paste, and concrete; and bricks (**Fig. 2**). The effects of various critical factors on the resulting products' performance, preparation mechanisms, leaching behavior, lead fates, and environmental and human safety were analyzed. The comparison of these recycling methods, and directions for future research, were discussed and reported as well.

2.1 Foam glass-ceramic

Because of its excellent intrinsic properties—such as low thermal conductivity, low water absorption and incombustibility—foam glass has attracted growing attentions, and has been applied in various fields, such as building and road construction, the petroleum and chemical industries, underground engineering, and military defense (Chen et al., 2012; Guo et al., 2010). From its physical aspect, foam glass is a porous thermal and acoustic-insulating material with high true porosity of up to 90-97%. It is a heterophase system, consisting of vitreous solid and gaseous phases. In the first phase, solid glass forms thin walls of single cells, which are filled during the second, gaseous, phase (Spiridonov and Orlova, 2003). Foam glass is generally produced with a powder method (Rawlings et al., 2006) consisting of mixing and sintering a mixture of glass cullet and foaming agents. When it is heated above the softening point, the solid glass becomes a viscous liquid, and the decomposition or oxidation of foaming agents lead to the formation of bubbles, which are trapped within the melt. The expanding gas bubbles increase the sample volume, thus forming a typical porous product (König et al., 2014).

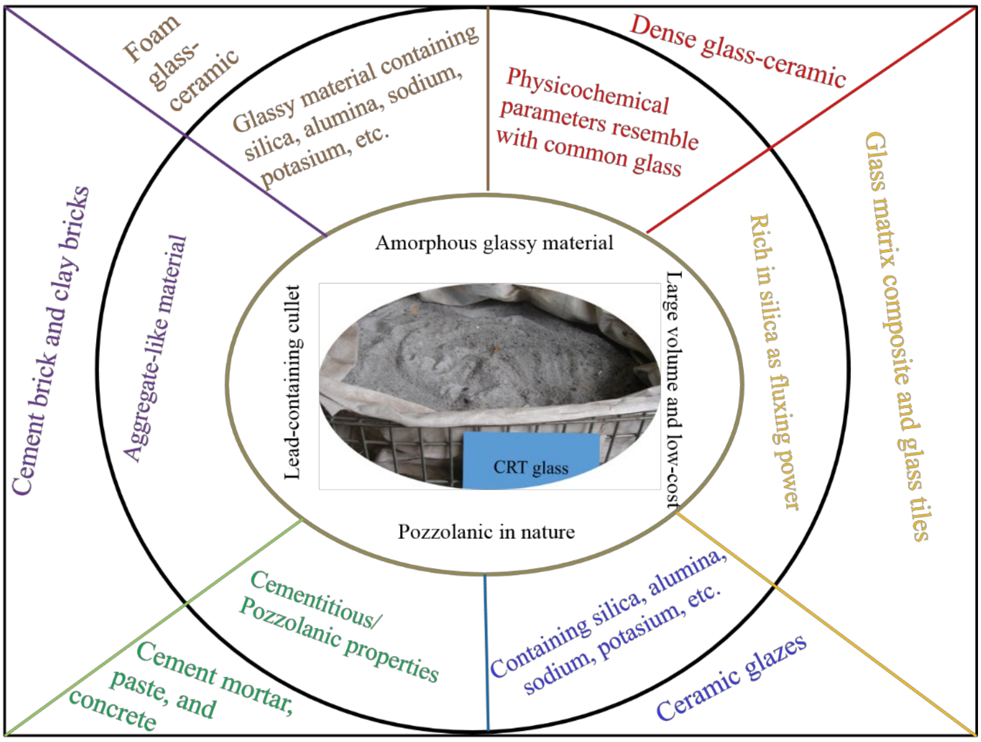

Fig. 2. Recycling CRT glass in the construction field

2.1.1 Effects of glass cullet

Foam glass is mainly produced from different types of glass cullets, such as flat glass, container glass, and cullet derived from discarded TV sets and computers, as well as the luminescent lamp glass. However, it can also be fabricated from other glassy materials, such as fly ash and slag. Recently, extensive studies have been carried out to use CRT glass (single funnel, panel glass or a mixture of these) for making foam glass (**Table 1**). Bernardo and Albertini (2006), König et al. (2015), Petersen et al. (2014), and Mucsi et al. (2013) fabricated foam glass by using CRT panel glass and different foaming agents (carbon, sodium carbonate and calcium carbonate). Guo et al. (2010) and Méar et al. (2006) used funnel glass with SiC and TiN as foaming agents to prepare foam glass. Mear et al. (2006; 2007) and Fernandes et al. (Fernandes et al., 2014) also prepared foam glass from a mixture of funnel and panel glass with SiC, TiN, egg shells, calcite and dolomite as foaming agents. The weight ratio of panel and funnel was found to affect the foaming behavior and consequently the product properties. Both panel and funnel glass consist of similar contents of modifier oxides (Na_2O+K_2O+CaO+MgO), whereas funnel glass presents a lower content of silica than of panel glass. In addition, funnel glass is rich in lead while panel is a barium-rich glass. The distinction between their chemical compositions results in different thermal behaviors. In fact, the glass should attain low enough viscosity (10^7-10^8 poise) for expanding by the gas released from the foaming agents under the internal pressure. Funnel glass was observed to be more prone to foam at lower temperatures, due to its lower refractoriness as compared with panel glass. It was reported that the sintering temperature for a panel glass and eggshell mixture (apparent density 0.38 g/cm^3 at 700°C) was about 50°C higher than that for a mixture comprising of funnel glass and eggshell to achieve a same foam density (Fernandes et al., 2013). The funnel glass underwent immediate expansion after the maximum shrinkage was attained due to the entrapment of CO_2/air inside the melt, while expansion was delayed for panel glass (Fernandes et al., 2014). Using funnel glass could enhance the foaming ability, whereas panel glass improved the compressive strength of the foam product. Regardless of the foaming agents (SiC or TiN), lower porosities and higher bulk density were obtained for panel glass, and the highest porosities and densities for cone glass (Mear et al., 2013). Panel-containing compositions obtained a higher mechanical strength than funnel-containing ones, even with lower apparent density values. Glass foams featuring apparent density and compressive strength of 0.29 g/cm^3 and 2.34 MPa, respectively, could be obtained from a mixture of both glasses in equal amounts, upon heat treating at 700 °C for 15 min using egg shells as a foaming agent (Fernandes et al., 2014). Benzerga et al. (2015) studied the effect of cullet glass (soda-lime silicate glass (SLS), CRT funnel glass or a mixture of the two) on the physical properties of foam glass. Different foam densities resulted from the cullet composition, and the values increased as the SLS fraction rose. Scanning electron microscopy observation showed that the distinction in foam density was reflected by an increase in the pore size. In fact, although the bulk density of CRT glass (2.85 g/cm^3) was higher than that of SLS glass (2.5 g/cm^3), the presence of lead made it more reactive to the foaming process.

2.1.2 Effects of foaming agents

It is generally recognized that foaming agents can be grouped into redox and neutralization agents (Fernandes et al., 2009; Spiridonov and Orlova, 2003). Redox foaming agents are usually carbon-containing materials (e.g., graphite, carbon black, silicon carbide (SiC) or organic compounds) and, less frequently, nitrides (e.g., titanium nitride (TiN), boron nitride (BN), aluminum nitride (AlN) and silicon nitride (Si_3N_4)). The gaseous emissions (e.g., CO_2, CO or N_2) from these materials are associated with an oxidation reaction that effectively uses the oxygen available from the oxides in the glass structure (**Eqs. 1-3**). However, Brusatin et al. (2004) stated that redox agents were not preferable for use as foaming agents for lead silicate glass, because they might interact with large amounts of dissolved oxygen in the lead silicate or with PbO, resulting in a lack of oxidative conditions. Regarding this concern, Bernardo et al. (2010) added MnO_2 as an "oxidation promoter" to provide extra oxygen. When using SiC as the foaming agent, MnO_2 operates in the oxidation reaction to release CO_2, as shown in **Eqs. 4** and **5**. Heydari et al. (2014) reported that Co_3O_4 performed better than Fe_2O_3 to improve SiC oxidation and increase the foam porosity. Fe-rich glass may also act as an oxidation promoter for redox agents, due to the Fe^{3+}/Fe^{2+} high temperature reduction (Chinnam et al., 2013).

$$2SiC(s) + 3O_2(g) \rightarrow 2SiO_2(s) + 2CO(g) \qquad (1)$$

$$SiC(s) + 2O_2(g) \rightarrow SiO_2(s) + CO_2(g) \qquad (2)$$

$$2TiN(s) + 2O_2(g) \rightarrow 2TiO_2(s) + N_2(g) \qquad (3)$$

$$2MnO_2(s) \rightarrow 2MnO(s) + O_2(g) \qquad (4)$$

$$SiC(s) + 4MnO_2(s) \rightarrow SiO_2(s) + CO_2(g) + 4MnO(s) \qquad (5)$$

The second group usually comprises carbonates (e.g., calcite, dolomite or ankerite) or sulphates, which decompose upon heating, with emissions of CO_2 or SOx (**Eqs. 6** and **7**). Intense gas release during their decomposition breaks the walls of individual pores, which merge and create a maze-like system of cavities in glass. It is worth noting that the reaction products—e.g., CaO from the carbonates' decomposition—remain in the bubble, possibly influencing glass properties such as viscosity and crystallization behavior. König et al. (2014) studied the influence of $CaCO_3$ content on foam density and found that the decomposed CaO was dissolved in the glass matrix. Petersen et al. (2014) also revealed that the collapse of foam at relatively high Na_2CO_3 content occurred due to the incorporation of Na_2O into the glass matrix. The presence of Na_2O in the silicate network provided non-bridging oxygens and thereby caused depolymerization of the primary $[SiO_4]$ network, and a decrease in the viscosity. Therefore, to avoid such adverse effect, redox agents are always preferred. Oxidation and decomposition may even overlap, as in the case of nitrides, being transformed into oxides and releasing N_2.

$$CaCO_3(s) \rightarrow CaO(s) + CO_2(g) \qquad (6)$$

$$CaMg(CO_3)_2(s) \rightarrow CaCO_3(s) + MgO(s) + CO_2(g) \qquad (7)$$

Generally, the foaming agents influence the foaming process: e.g., the foaming temperature and gas volume. Méar et al. (2006; 2007) investigated the effects of SiC and TiN on the microstructure evolution of foam glass. The bursting of ‘‘bubbles’’ by the gas emissions (N_2 or CO_2) created pores in the expanded sample. Macropores with a single distribution were formed after reduction with SiC, whereas a double pore size distribution was observed for TiN agents. Benzerga et al. (2015) studied the influence of foaming agents (C, SiC or AlN) on the foam density, and AlN was found to be a more efficient foaming agent than SiC. The result of Fernandes et al. (2014) showed that using calcite as the foaming agent featured lower apparent density regardless of the glass type, and the highest density value was obtained for dolomite-containing foams from panel glass. Since the attained viscosity at a certain time-temperature condition was the same for compositions containing a given glass, the dominant factor for the foaming process will be the decomposition behavior of the calcite, dolomite and eggshell.

The foaming agent contents also have an effect on the foam products’ performance. Fernandes et al. (2013) studied the effect of eggshell content (0-5 wt.%) on glass foaming ability via sintering mixtures of CRT glass and eggshell at 700 °C for 15 min. The apparent density decreased as the eggshell content increased, and stabilized at 0.3-0.4 g/cm^3 with the inclusion of 3-5 wt% eggshell. König et al. (2014) found that with 1-2 wt% $CaCO_3$ exhibited a dense sintered glass shell around a foamed core, indicating that $CaCO_3$ was decomposed before the glass particles sintered and closed the porous structure. The shell gradually became thinner and ultimately no longer visible for the sample with 4 wt% $CaCO_3$. For samples with 10 wt% $CaCO_3$, the porous structure did not close completely and the released gases were able to escape. Petersen et al. (2014) reported that with small amounts of Na_2CO_3 (2-10 wt%) as foaming agent the specimens exhibited regular shapes regardless of their foaming temperature. However, samples foamed with greater amount (14-22 wt%) of Na_2CO_3 exhibited a temperature dependent sample shape.

2.1.3 Effects of foaming temperature and time

Glass viscosity, foaming temperature and residence time are strongly correlated. If the foaming temperature is high, the melt viscosity will be low, and controlling the structure will become difficult because bubbles rise to the top of a mold. Conversely, if the temperature is low and possess a higher glass viscosity, then the gas expansion becomes difficult and little increase in volume occurs (König et al., 2014; Mucsi et al., 2013; Petersen et al., 2014). König et al. (2014) studied the effects of foaming temperature and time on the density, porosity and homogeneity of foam glass. When the foaming temperature was increased or decreased by 30 °C from 785 °C for the 5-

min-foamed samples, the apparent density increased by 15-20%. The increase in the densities with foaming temperature higher than the optimum was related to the collapse of foam, resulting from the decrease of viscosity and secondary effects, i.e. pore coalescence and pore opening. Prolonging foaming time from 5 to 15 min at 785 °C resulted in the apparent density's being maintained at the same level. With a further increase in the time to 30 min, a drastic increase in density was observed. Petersen et al. (2014) reported a minimum density of 0.28 g/cm^3 when 14 wt% Na_2CO_3 was added into CRT panel glass at foaming temperature of 750 °C. At lower temperatures (700-750 °C), the samples had a dense outer rim with a hollow interior, and at higher temperatures (850-900 °C), the samples were mostly collapsed. Méar et al. (2006; 2007) investigated the influence of reaction time and temperature on the microstructure evolution and mechanical behavior of foam glass. Increasing reaction temperature and time could increase the size of pores and the heterogeneity of their distributions in the foam glass, thus reduced the mechanical resistance. König et al. (2015) studied foaming conditions' influence on the density and homogeneity of foam glass. At lower temperatures, the reaction was slower and the amount of gas released was less, while at higher temperatures, the pores grew faster, coalesced and became open, leading to the collapse of foam. The combination of a large MnO_2 amount and high temperature strongly accelerated the collapse of foam. Mucsi et al. (2013) prepared foam glass using CRT panel glass, limestone and dolomite as foaming agents. The influences of sintering-condition studies indicated that temperature had no significant effect on the density of pellets at 600-750 °C regardless of the residence time. A significant decrease in particle density was observed at 800 °C for 7.5- and 10-minute durations; this decrease occurred at 850 and 900 °C even for a 5-min residence time.

2.1.4 Effects of lead in glass cullet

Foam glass manufacturing is a promising recycling mode for CRT glass. Nevertheless, because CRTs employ glass containing toxic metals, the development and leaching behavior of the fabricated products is therefore important. Méar et al. (2005; 2006) studied the reaction of foaming agents (SiC and TiN) between PbO. XRD analysis of the foam glass revealed the formation of Pb^0. Energy dispersive spectroscopy analysis indicated that the bubbles on the pore surface could be ascribed to the lead formed during the reaction process as a result of lead oxide reduction (**Eqs. 8** and **9**). The Pb^0 amount increased with the inclusion of foaming agents; the increasing trend was exponential for TiN and linear for SiC. The remaining lead oxide accounted for nearly 75% of the lead in the raw CRT glass. X-ray photoelectron spectroscopy analysis revealed that the remaining lead oxide was in the form of $PbSiO_3$, a lead oxide in a silicate environment.

$$PbO(s) + SiC(s) \rightarrow Pb(s) + SiO_2(s) + CO_2(g) \quad (8)$$

$$PbO(s) + TiN(s) \rightarrow Pb(s) + TiO_2(s) + N_2(g) \quad (9)$$

The work of Guo et al. (2010) and Benzerga et al. (2015) also revealed that the reaction of PbO and SiC generated Pb^0. Small white dots in SEM images and XRD analysis confirmed the presence of Pb^0 microcrystal. Yot and Méar (2011) studied the leaching behavior of lead, barium and strontium from foam glass and found that the Pb^{2+} concentrations were always higher in leachates (□110mg/L) for foam prepared using TiN compared to the case of SiC (□3mg/L), irrespective of the glass powder (funnel, panel, or mixture) used. Most samples yielded leaching rates below the regulatory limit of 5 mg/L (according to U.S. EPA and China MEP., Chen et al., 2009a), while the foam obtained using funnel glass with 4 wt% TiN exceeded the legislative limit. This was attributed that more Pb^0 was formed by lead reduction inside the glassy framework when TiN was employed. The levels of Ba^{2+} released (0.60~40.40 mg/L) were under the regulatory limit of 100 mg/L regardless of the foaming-agent content. The amount of Sr^{2+} released (5.20~51.30 mg/L vs. 1.70~6.90 mg/L for pure original glass) depended on the inclusion of funnel glass and the foaming agent content.

For foam glass, the metals present in CRT glass were generally transferred to the regenerated products but not removed or separated; hence the potential threat remained. To detoxify the leaded glass, Chen et al. (2009b) developed pyrovacuum reduction to recover lead from CRT funnel glass and synchronously transferred the residue into foam glass. In this process, the lead oxide was first reduced to lead by carbon, then the lead evaporated into a gaseous phase and was recovered after cooling (**Eq. 10**).

$$PbO(s) + C(s) \rightarrow CO(g) + Pb(g) \quad (10)$$

Chen et al. (2009a) used self-propagating process to detoxify CRT glass. During the treatment, SiO_2 was partially released from glass network and Pb played a role of glass former (**Eq. 11**). These effects resulted in an increase in Si-O-Pb linkage and decrease in Si-O-Si linkage. Therefore, it is more difficult for lead to leach from the final products than to leach from the original glass.

$$SiO_2 + 3Mg^o + Fe_2O_3 = Mg_2SiO_4 + MgO + 2Fe^o \quad (11)$$

Nevertheless, a possible barrier to using CRT glass in these applications is the potential threat to human health associated with lead-containing products. Innovative technologies have been developed to detoxify the leaded glass, include the mechanochemical processes (Yuan et al., 2012), ultrasonically enhanced leaching (Saterlay et al., 2001), subcritical water-aided leaching (Miyoshi et al., 2004), self-propagating methods (Chen et al., 2009a) and reduction-melting processes (Okada and Yonezawa, 2014). Saterlay et al. (2001) used ultrasound to facilitate lead leaching from CRT glass, achieving a removal rate of over 90% of the leachable lead. Lu et al. (2013) recovered lead from CRT funnel glass by thermal reduction with metallic iron, and 58 wt% lead extraction was achieved. Yuan et al. (2012) applied mechanical activation to pretreat CRT funnel glass, followed by diluted nitric acid leaching, and a high yield of 92.5% of the lead was achieved. Erzat and Zhang (2014) used chloride volatilization to recover lead from CRT funnel glass. From the above literatures, it can be seen that lead can be effectively removed from leaded glass using recent advanced technologies.

Table 1. Representative foam glass-ceramics derived from CRT glass

CRT glass	Foaming agents	Foaming conditions	Porosity (%)	Apparent density (g/cm^3)	References
Single funnel glass, panel glass or a mixture	SiC or TiN	850°C for 1 h	3.7-86.1	—	(Yot and Méar, 2011)
Single funnel glass, panel glass or a mixture	SiC or TiN and oxide agent MgO	800°C for 1 h	65.1-80.6	0.38-1.35	(Mear et al., 2006; Méar et al., 2007)
Panel glass	Carbon black	780-830°C for 10-50 min	—	—	(Lian, 2012)
Funnel glass	SiC or TiN	750-950°C for 30-90 min	—	—	(Méar et al., 2006)
Single funnel glass, panel glass or a mixture	Eggshell, calcite or dolomite	650-750°C for 15 min	—	0.27-0.49	(Fernandes et al., 2014)
Panel glass	Carbon and MnO_2	780-840°C for 5-60 min	—	—	(König et al., 2015)
Panel glass	Na_2CO_3	700-900°C for 45 min	—	0.28	(Petersen et al., 2014)
Panel glass	$CaCO_3$	725°C for 5-30 min	—	0.07-0.12	(Bernardo and Albertini, 2006)
Panel glass	SiC, oxidation	850 and	34.4-52	—	(Saeedi

	agents Fe_2O_3 or Co_3O_4	1050°C for 105 min			Heydari et al., 2014)
Panel glass	$CaCO_3$	755-815°C for 5-30 min	90.2-91.2	0.24-0.27	(König et al., 2014)
Panel glass	Dolomite and limestone	600-900°C for 5-10 min	—	0.6-0.9	(Mucsi et al., 2013)
Panel glass	SiC, flux agent sodium borate, and stabilizer TiO_2	860-930°C for 30 min	—	0.20-0.23	(Zhang et al., 2016)
Single panel or funnel glass	Egg shell	600-850°C for 15 min	—	0.28-0.90	(Fernandes et al., 2013)
Panel glass	C, AlN or SiC	850°C for 30 min	—	0.36-0.77	(Benzerga et al., 2015)

2.2 Dense glass-ceramic

For dense glass-ceramic preparation, fine glasses were generally pressed and sintered, to allow the crystallization occurring together with densification (**Table 2**). Bernardo et al. (2006; 2007) employed CRT panel glass, lime and mining residue to prepare wollastonite- and sanidine-based sintered glass-ceramics. The mixture was subjected to sintering at low temperatures (880-930 °C) with concurrent crystallization. The achieved mechanical properties (bending strength>100 MPa, Vickers micro-hardness>7 GPa) together with the simplicity of the manufacturing method, make it promising for use as a construction material. Ponsot et al. (2013) used CRT panel glass, exhausted lime and kaolin to fabricate sintered glass-ceramics. The starting mixture was pressed to form discs or rectangular tiles. The molded specimens were first sintered at 800-1100 °C, and then the disc specimens were removed from the furnace after 30 min of holding time, while the tiles were cooled at the end of the holding stage. Sintered glass-ceramics featured a water absorption below 2%, good strength and elastic modulus. Eftimie and Melinescu (2015) prepared glass-ceramics using a mixture of CRT glass (weight ratio of neck, panel and funnel=5:30:65) and TiO_2 as the nucleating agent. The raw materials were first shaped using cold pressing, then sintered at 700-800 °C for 30 min. A good stability and decrease of thermal expansion coefficient (TEC) were observed for samples with 5 wt% TiO_2. X-ray diffraction revealed the presence of anorthite along with a large amount of vitreous phase. Reben et al. (2015) also added nucleating agents ZrO_2 and TiO_2 (8 and 15 wt%) to panel glass to obtain fresnoite glass-ceramics via surface crystallization. TiO_2 was found effective in decreasing the crystallization temperature, whereas ZrO_2 increased it. The TEC decreased with an increase in ZrO_2 content. However, only TiO_2 in the CRT glass led to the surface crystallization of fresnoite. Eftmie and Ţacu (2014) used a mixture of CRT glass (5% neck, 30% funnel and 65% panel) and ZrO_2 to prepare glass-ceramics. Andreola et al. (2005; 2010) mixed panel or funnel glass with different amounts of dolomite and alumina to favor the crystallization process or to improve the chemical resistance and hardness of the glass. Glass-ceramics composed of Nepheline, Akermanite and Celsian were obtained at low temperature and in a short time (900 °C for 60 min). Although the Pb has low field strength and large radius comparable to those of Ba and Sr in the panel glass. However, the crystallization capacity of the funnel-containing compositions was higher than that of panel-containing ones. Pb-containing crystalline phase had also not been detected, confirming that Pb remained in the glass matrix and was not involved in the devitrification process.

Table 2. Representative dense glass-ceramics derived from CRT glass

CRT glass	Additives	Sintering conditions	Crystalline phases	References
Panel glass	Mining residues and lime	880-930°C for 0-5 h	Wollastonite, sanidine, albite and trikalsilite	(Bernardo et al., 2007)
Panel glass	Mining residues and lime	880°C for 0-3 h	Sanidine, trikalsilite, panunzite, Ca alumino-silicate, and Ca-K silicate	(Bernardo et al., 2006)
Panel glass	Exhausted lime and kaolin clay	800-1100°C for 30 min	Wollastonite, pseudowollastonite, cuspidine, and anorthite	(Ponsot et al., 2013)
Mixture of neck, funnel and panel glass	TiO_2	700-800°C for 30 min	Anorthite	(Eftimie and Melinescu, 2015)
Panel glass	ZrO_2, TiO_2 or a mixture	Melted at 1450°C for 2 h, annealed at 500-600°C, and heated at 800°C for 24 h	Fresnoite	(Reben et al., 2015)

2.3 Glass matrix composite and glass tiles

Silicate glasses have exceptional optical, thermal, and mechanical properties, allowing technical applications in the fields of optics, microelectronics and chemical technology. However, the limitation of brittleness does not endow them to be reliable structural materials. One solution to reinforce glass is incorporating reinforcements and forming a glass matrix composite. Minay et al. (2003) proposed an extrusion technique to fabricate glass matrix composites through reinforcing CRT panel glass with Al_2O_3

platelets. The glass powder and Al_2O_3 platelets were mixed and pressed to obtain pellets. The extrusion was carried out at 700 °C with a holding time of 15-30 min. Bernardo et al. (2003; 2004; 2005; 2007; 2009) employed a CRT glass mixture to manufacture alumina platelet-reinforced glass matrix composites for tile applications. Notable bending strength values (>105 MPa) were achieved for composite sintered at 650 °C for 15 min.

The reuse of CRT glass in the fabrication of porcelain stoneware tiles has been investigated by Tucci et al. (2003), who highlighted the strong fluxing power of leaded glass, being able to improve the formation of both liquid phase and mullite when present in a low amount (<2 wt.%). Raimondo et al. (2007) also studied the effects of glass additions (5 and 10 wt.%) on the sintering behavior of porcelain stoneware tiles. These glasses could partially replace conventional flux feldspar without significantly affecting the technological process. However, some PbO was lost during firing (0.2-0.3 wt%) and a small amount after firing (<0.7 mg/kg). Souza et al. (2004) prepared porcelain stoneware tile using mixes containing funnel glass as a partial replacement of Na-feldspathic. Use of lead-containing glass had little effect on the microstructure compared with standard composition. The presence of PbO in the composition of stoneware enhanced vitrification of the body mix at earlier stages of firing. Chemical analysis of the as-fired samples revealed that some PbO had evaporated.

2.4 Ceramic glazes

Ceramic glazes have essentially two functions: technical and aesthetic. The former is to render the ceramics surface completely waterproof and the second is to give ceramics a glossy and colorful surface (Schabbach et al., 2011). Lazău et al. (2013) prepared frits by melting CRT panel glass and raw materials at 1,250 °C with a soaking time of 30 min. The frits were then used to prepare ceramic glazes containing 95% frit and 5% kaolin. Andreola et al. (2005; 2007) investigated the substitution of CRT glass (panel and funnel) for common ceramic frits in glazes manufacture. The investigation, undertaken in both the laboratory and industry, found that the obtained glazes had similar aesthetic and mechanical properties as standard glazes. Life cycle assessment (LCA) of the standard and CRT glass glaze indicated that the production of CRT glass glaze led to an overall reduction of environmental damage by 36%. Siikamäki (2006) also reported that panel glass was a suitable raw material for ceramic glazes. The properties of glazes containing up to 14.5 wt% of CRT glass were similar to commercial glazes. Schabbach et al. (2011) prepared ceramic glaze by using CRT cone glass and pre-treated incinerator bottom ash. The reformulated glazes showed better acid resistance and aesthetic characteristics. Leaching tests indicated that the resulting glazes showed a significantly lower lead release.

2.5 Cement mortar, paste, and concrete

2.5.1 Cement mortar and paste

Natural river sand is commonly used as a fine aggregate in cement mortar production. However, excessive excavation of sand causes serious environmental problems (Sua-Iam and Makul, 2013). The reuse of CRT glass to replace river sand and cement in concrete is feasible because of its identical chemical structure. Zhao (2013) and Zhao and Wei (2011) reported the partial substitution of untreated and nitric acid-treated CRT glass for natural sand in mortar mixtures. An increase in workability was observed for the fresh mortar. The initial slump of concrete increased by 112.5, 200 and 237.5% with the inclusion of 25, 50 and 75% acid-treated CRT glass, respectively. Similar finding was also reported by Ling and Poon (2012; 2012a, 2013) indicated that the inclusion of glass cullet improved the fluidity, a characteristic that may be attributed was the lower water absorption and the smooth and impermeable surfaces of the CRT glass used. Therefore, the use CRT glass could reduce the usage of chemical admixtures, such as superplasticizer or water reducer for achieving a same workability of mortars. Ling and Poon (Ling and Poon, 2013) further revealed that the workability of CRT mixtures also depended on the particle size of the glass cullet used. Glasses with a maximum size of 5 or 2.36 mm performed better in the workability than those smaller particle sizes of 1.18 mm or 0.6 mm.

Alkali-silica reaction (ASR) in cement is a major contributor to the failure of cement structures, causing increased repair costs and possible rebuilding expenses. Ling and Poon (2011) found that an increase in the amount of CRT glass in cement mortars led to an increase in ASR expansion—a shortcoming of this recycled material (Rashad, 2014). However, the ASR expansion can be successfully mitigated by adding supplementary cementitious materials, such as fly ash (Ling and Poon, 2011), lithium additives (Demir and Arslan, 2013), slag (Thomas and Innis, 1998), metakaolin (Gruber et al., 2001), silica fume (Shi and Zheng, 2007), etc.

The strength of mortar is also commonly considered a valuable property, and it usually gives a good overall picture of the mortar quality. Ling and Poon (2014) replaced the recycled fine aggregate in non-load-bearing (NLCB) and load-bearing (LCB) concrete blocks with treated funnel glass. The compressive strength of NLCB at 28 days decreased from 22.3, to 18.4 and 15.3 MPa, as the glass content increased from 0, to 50 and 100%, respectively. It was evident that the inclusion of CRT glass in the matrix decreased the strength associated to the weak adhesion between the smooth surfaces of CRT glass and the cement paste interface (Zhao et al., 2013). With an increase in the amount of CRT glass, more residual lead derived from CRT glass was added into cement, which retarded the cement hydration and inhibited cement hydration product formation. However, some studies reported the mortars with CRT glass could slightly increase the strength. Zhao et al. (2013) related the higher strength was due to the improvement of aggregate packing in the mortar system. In addition, the very fine CRT glass particles can react as filler or pozzolan to accelerate the hydration of cement. Maschio et al. (2013) prepared high-strength mortar with the use of milled CRT panel glass and superplasticizer. Specimens containing CRT glass showed a more rapid increase in strength with respect to the reference compositions. Moncea et al. (2013) used panel and funnel glass (over 95% funnel glass) as supplementary cementitious material in mortars based on Portland cement and slag cement, as well as for the partial replacement of the solid component in alkali-activated slag/fly ash binders. The compressive strengths of studied mortars were almost identical to reference mortar. The differences in the reported values of compressive strength become a deterrent to the use of CRT glass in mortar preparation. However, it has been reported by Ling and Poon (2014) that the way to overcome this problem is by maintaining a proper aggregate-to-cement ratio and using appropriate casting methods.

Shrinkage of mortar is an increasingly important issue, as an improper shrinkage can lead to cracking and poor serviceability. Zhao et al. (2013) reported a decrease in drying shrinkage of high-density concrete containing nitric acid-treated CRT funnel glass. This reduction increased with an increase in glass content; the reductions were 3.2, 10.28 and 13.35% with the inclusion of 25, 50 and 75% glass cullet, respectively. Ling and Poon (2012b) also reported a reduction in the drying shrinkage—at ages of 56 and 112 days for barite concretes containing CRT glass. A study by Ling and Poon (2014) revealed that CRT glass decreased the total water content and thereby reduced the shrinkage. The reduction in the drying shrinkage shows the advantage of using this recycled material. However, some researchers reported a negative effect of CRT glass inclusion on the drying shrinkage.

The leaching characteristics of cement mortar containing CRT glass were studied by Ling and Poon (2012; 2012a, 2013), and found that the lead leaching from mortar samples prepared with acid-treated CRT glass complied with regulatory limits. The report of Moncea et al. (2013) revealed that the cumulative lead released in leachates after 64 days for mortars including CRT glass was below the emissions limit. To assess the effect of acid-treated funnel glass content on the lead leaching of the concrete blocks, Ling and Poon tested samples with a 25% replacement level of recycled fine aggregate of CRT glass (Ling and Poon, 2014). The results indicated that the lead leaching was below the TCLP limit. They also assessed the influence of casting methods on the variability in lead leaching levels for concrete blocks. By removing the manual compaction during the casting process, the lead leaching of samples with a replacement by volume of 100% was significantly reduced, from 27.44 and 9.77 mg/L to 4.75 and 3.79 mg/L, respectively. This is because the manual compaction applied during the casting process could break the glass easily and results in a significant leaching of lead from the broken glass.

2.5.2 Concrete

CRT glass is rich in silica and pozzolanic in nature, making it as a potential substitution for river sand in concrete. Romero et al. (2013) used CRT glass as a fine aggregate replacement in concrete. Durability, strength and leaching tests were conducted to investigate the comprehensive performance of CRT-concrete. The results indicated that the strength of CRT-concrete exceeded that of the control sample. However, the CRT-concrete was susceptible to ASR expansion if more than 10% glass cullet was included in the mixture. Leaching tests showed that the lead concentration of CRT-concrete could be below the drinking-water limit, but this effect depended on the glass content and whether biopolymers were used. Walczak et al. (2015) also revealed that the use of CRT glass resulted in a 16% increase in the compressive strength, and a 14% increase in the flexural strength, of concrete mortar. Sua-iam and Makul (2013) partially replaced the natural sand in self-compacting concrete mixtures with CRT glass. A reduction in the slump flow was observed, and the initial and final setting times increased with an increase in CRT glass content. Ling and Poon (2014) used acid-treated funnel glass to replace fine aggregate in concrete blocks. All the blocks demonstrated acceptable compressive strength and ASR expansion, but also improved resistance to water absorption and drying shrinkage. To limit the possible leaching of lead, it is best to limit the inclusion of CRT glass in concrete blocks to below 25%.

Heavy-weight concrete is one of the most common types of concrete used in nuclear power plants, medical units and other structures where radioactive protection is required. Ling and Poon (2012b) investigated the feasibility of using untreated and acid-treated CRT funnel glass as partial and full replacements of fine aggregates in heavyweight barite concrete. The overall properties of the obtained barite concrete were comparable, except for the lead-leaching results. Although it was feasible to use the treated CRT glass as 100% substitution of fine aggregate, they found that the inclusion of CRT glass in concrete should be controlled below 25%, to decrease the possibility of lead leaching. Tian et al. (2016) fabricated anti-radioactive concrete using CRT funnel glass to replace both fine and coarse aggregate, and the optimum percentage of funnel glass used as either fine or coarse aggregate was determined to be 40%. The use of CRT glass considerably improved the radiation shielding performance.

2.5.3 Leaching behavior

In a cementitious system, such as cement mortar and concrete, the release of alkalis and hydroxyl during the cement hydration process could yield a high pH value of 11-13 (Tariq and Yanful, 2013). The cement's capacity of encapsulating pollutants in the hardening structure as well as the high pH value of intergranular solution, which favor the formation of metal hydroxide, along with the existence of calcium silicate hydrates (C-S-H) with high specific surface areas, are key factors of toxic metals immobilization in cementitious system (BĂDĂNOIU et al., 2008; Georgescu et al., 2006). In order to investigate the environmental compatibility of these materials, toxicity characteristics leaching procedure (TCLP) were carried out for the determination of lead leaching (Iniaghe and Adie, 2015). The results showed that concrete matrix was better suited than cement mortar for recycling CRT funnel glass. The greater volume of natural fine aggregates in concrete may be responsible for the observed drastic reduction of leachable lead. However, the extent of lead leaching was significantly greater than the regulatory limit of 5 mg/L, when the CRT glass replacement level was greater than 25%. Cement usually contains oxides of Ca, Si, Al, and Fe, which can form hydrogen bonds with a biopolymer's amino, hydroxyl, carbonyl, and carboxylic repetitive groups during hydration. Based on this, toxic meals could be entrapped in the biopolymer-modified concrete system (Kim et al., 2012).

2.6 Cement brick and clay brick

The manufacturing procedure for bricks in displayed in **Fig. 3**. Lee et al. (2012; 2016) used CRT panel glass as an aggregate in concrete and clay bricks. Both types of bricks containing panel glass met the Korean Standards KS F 4419 and KS L 4201, respectively, but the flexural strength for concrete bricks and compressive strength for clay bricks decreased as glass content increased. A maximum of 40% panel glass can be incorporated into concrete blocks, and about 2% funnel glass into clay bricks. Andreola et al. (2010) also added CRT panel glass to a commercial brick body for external facing bricks, in amounts ranging from 0.5 to 3.0 wt%. The glass was not involved in the solid-state reactions during firing. Its properties were in accordance with the industrial tolerance values regarding shrinkage, weight-loss and flexural-strength values. A study by Dondi et al. (2009) revealed that the recycling of both funnel and panel glass into clay bodies was technically feasible. Although no significant release of Pb, Ba, or Sr was observed during the firing and leaching tests for carbonate-poor bodies, but some Pb volatilization during firing and Sr leaching were observed for carbonate-rich bodies. The recommended amount of CRT glass was within the range of 2-4%, depending on the characteristics of the clay bodies.

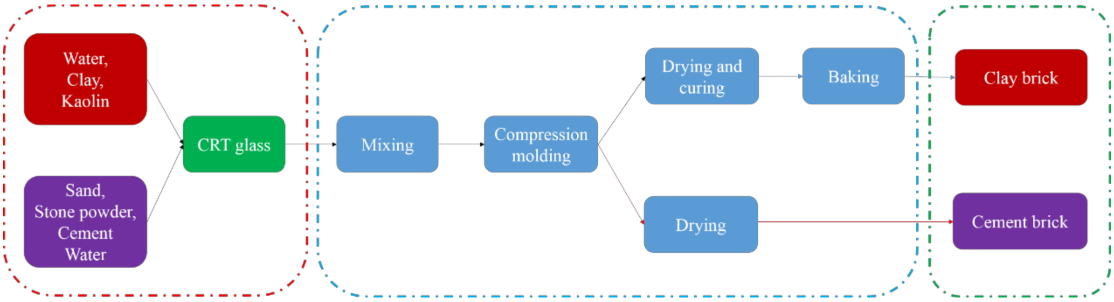

Fig. 3. Manufacturing procedure for bricks (Lee et al., 2016)

3. Conclusions

Although there has been steady progress in these recycling possibilities in recent years, there are still some limitations on these current applications. The notice and improvement are thus proposed **(Table 3)**. Fabricating foam glass-ceramics offers advantages of saving limited natural resources by partially replacing common glass cullet and cutting waste disposal costs. Many factors are reported to affect the foam products' performance, including the characteristics of the glass cullet, the foaming agents and the foaming conditions. Leaded glass is more prone to foam at lower temperatures, because of its lower refractoriness. However, metallic lead is formed on the pore surface of foam as a result of lead oxide reduction. The leaching rate of foams mainly depends on the inclusion of leaded glass and the foaming agent, and most of the studies reported that the leached lead is below the regulatory limits. For the preparation of dense glass-ceramics, the inclusion of CRT glass can favor the crystallization process. The nucleating agents used affect the glass-ceramic performance, including the water absorption, strength, TEC and crystallization temperature. The lead remained in the glass matrix, even though no lead-containing crystalline phase was detected. Regarding the fabrication of glass matrix composite and glass tiles, CRT glass could partially replace common fluxing agents and enhance vitrification of the body mix at early stages of firing. However, the evaporation of lead during firing is inevitable, and strict pollution control is needed. For the preparation of ceramic-glazes, CRT glass could partially replace common frits, and the glaze products display good aesthetic and mechanical properties. However, because some lead remains in the glazes, further studies are needed, to determine how to minimize the lead content. Preparing cement mortar, paste, and concrete from CRT glass offer added advantages in terms of quantity of recyclable cathode ray tube glass at a given time, with minimal environmental and economic implications. With significant quantity of CRT glass being generated globally, cementitious systems could be economically and environmentally acceptable as a sound management practice for CRT glass.

Table 3. Comparison of different applications of CRT glass

Applications	Advantages	Disadvantages/Limitations	Notice/improvement
Foam glass-ceramic	1. Partially replaces common glass cullets, and thus saves limited natural resources. 2. Turns an otherwise useless waste into products and cuts waste disposal costs. 3. Leaded glass is more prone to foam at lower temperatures, and thus reduces energy consumption.	1. Various factors have effects on the foam products' performance, including the types of CRT glass and the inclusion amounts. 2. Metallic lead may form on the pore surface of foam as a result of lead reduction.	1. Reasonably controls the inclusion of leaded glass. 2. Further studies needed, of the various factors affecting the products' performance and the leaching characteristics of foam products.
Dense glass-ceramic	1. Partially replaces common glass cullets, and thus saves limited natural resources. 2. Favors the crystallization process and/or improves the chemical resistance of glass.	1. Lead remains in the glass matrix. 2. The nucleating agents used affect the glass-ceramic performance.	1. Reasonably controls the inclusion of leaded glass. 2. Further studies needed, of the environmental and human safety of glass-ceramic products.
Glass matrix composite and glass tiles	1. Partially replaces common fluxing agents, and thus saves limited natural resources. 2. Enhances vitrification of the body mix at early stages of firing.	Some lead is lost during firing.	1. Reasonably controls the inclusion of leaded glass. 2. Strict pollution control needed because of the evaporation of lead during firing.
Ceramic glazes	1. Partially replaces common ceramic frits, and thus saves limited natural resources. 2. The glaze products display	The lead remains in the glazes, although the leaching rate is almost below the regulatory limit.	Further studies needed, of the preparation techniques for minimizing lead content by reformulating new glazes.

	good aesthetic and mechanical properties.		
Cement mortar, paste and concrete	1. Partially replaces fine aggregate, and thus reduces the environmental impact of excessive excavation of river sand. 2. Reduces production costs of cement mortar and concrete; decreases greenhouse gas emissions during cement production process. 3. Enhances the fluidity and workability of fresh concrete; improves the radiation shielding performance of concrete.	1. Increases the ASR expansion of concrete. 2. Possibly reduces the comprehensive strength of concrete and results in higher drying shrinkage.	1. Reasonably controls the inclusion of leaded glass. Adding biopolymers could prevent the possible leaching of lead. 2. The ASR expansion can be mitigated by adding cementitious materials, such as fly ash, lithium additives, slag, metakaolin, etc. 3. Further studies needed, of effects of CRT glass inclusion on the comprehensive strength and drying shrinkage of concrete. 4. The environmental and economic benefits depends on the end uses and production scale.
Cement brick and clay brick	Partially replaces aggregates, and thus reduces the environmental impacts of excessive excavation of sand, stone or kaoline.	Some lead is lost during firing.	1. Reasonably controls the inclusion of leaded glass. 2. Further studies needed, of the environmental and human safety of brick products.

4. Future research and prospects

Recent economic growth and the consequent rise in living standards have led to a drastic increase in large-scale construction and building projects, and a consequent shortage of resources and severe environmental impacts. The application of low-cost CRT glass in construction fields could partially replace some other common raw materials, and thus reduce the environmental impacts of excessive exploitation of natural resources. However, to the best of our knowledge, most reported technologies are still at the laboratory scale or in the early stages of commercialization. Further studies are needed to advance this research to the level of viable industrial application.

(1) The geographical distance between waste CRT glass sources (CRT recycling plants, CRT glass collecting groups, etc.) and processing facilities should be taken into account to yield maximum environmental and economic benefits. Generally, the raw materials should be transported no more than 100 km, otherwise the production costs of recovery will be prohibitive. The generation and recycling of waste glass in a bio-industrial park is one of best available practices.

(2) Commercial uses should be developed for recycled waste CRT glass, especially leaded glass. Although the recycling methods discussed in this study are technically feasible, there are significant differences in the volume of waste glass that can be used, for the various recycling methods and types of glass cullet. The amount of glass that can be used in the production of cement mortar, paste and concrete is relatively large, because the glass-containing products can easily meet the quality and leaching standards. The amount of leaded glass that can be used in these applications, however, is significantly less, because of the high lead levels in the glass. Further research is needed, therefore, to develop technologies that can incorporate more CRT glass into commercial products, and to minimize the amount of lead that leaches out. In addition, more uses are being developed, for mixtures of neck, funnel and panel glass, obviating to some degree the necessity of absolute separation of lead-free and lead-containing glass. Some practices have also been conducted, e.g. Hong Kong Environmental Protection Department collaborated with Hong Kong Polytechnic University and included CRT glasses in the regenerated concrete.

(3) Technology alone cannot make CRT glass recycling commercially viable; public policies must also promote such industries. State and local governments should issue policies that encourage glass recycling, such as reducing the effective tax rate for glass-recycling companies and preferentially purchasing recycled-glass products themselves. They could also conduct educational campaigns: for example, helping consumers understand that lead-containing products are safe when the amount of lead leached is below the regulatory limit. Due to the high level of lead in the CRT funnel glass, it is regulated under hazardous waste management regulations, which need be taken into account, especially for enterprises nonqualified for hazardous waste treatment.

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