



Water-soluble lead in cathode ray tube funnel glass melted in a reductive atmosphere



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HIGHLIGHTS

- Water-soluble lead species in glass after reduction-melting were identified.
- Metallic lead remaining in the glass dissolved in alkalized water.
- The residual metallic lead was regarded as water-soluble lead.
- The quantity of water-soluble lead was decreased by controlling melting conditions.

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ABSTRACT

In the reduction-melting process, lead can be recovered from cathode ray tube funnel glass (PbO = 25 wt%); however, resulting glass residues still contain approximately 1–2 wt% of unrecovered lead. For environmental protection in the residue disposal or recycling, it is important to evaluate the quantities of water-soluble species among the unrecovered lead. This study examined water-soluble lead species generated in the reduction-melting process of the funnel glass and factors determining their generation. In the reduction-melting, metallic lead was generated by reducing lead oxides in the glass, and a part of the metallic lead remained in the glass residue. Such unrecovered metallic lead can dissolve in water depending on its pH level and was regarded as water-soluble lead. When 10 g Na₂CO₃ was added to 20 g funnel glass during reduction-melting, the resulting glass contained high concentrations of sodium. In a water leaching of the glass, the obtained leachate was alkalized by the sodium-rich glass (pH = 12.7–13.0). The unrecovered metallic lead in the glass was extracted in the alkalized leachate. The quantity of the unrecovered metallic lead (water-soluble lead) in the glass decreased when the melting time, melting temperature, and carbon dosage were controlled during reduction-melting.

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1. Introduction

The recycling of waste cathode ray tube (CRT) glasses to produce new CRTs for televisions has been discontinued along with the manufacturing of CRTs for televisions. However, an increase in the waste CRT glass cullet between 2010 and 2020 in Asia has been reported [1]. Hence, an alternative technique for recycling waste CRT glass is necessary for resource recovery and waste minimization. Panel glass in CRTs can be converted to new products other than CRTs [2–7], whereas this direct conversion is difficult for funnel glass in CRTs because of the toxic lead oxides present in the

glass [8]. Therefore, it is necessary to recover the lead oxides from the funnel glass before converting the glass into new products.

Several researchers have proposed lead recovery techniques [9–20], and this study focuses on the reduction-melting process among the techniques. In this process, the lead oxides in the funnel glass are converted to metallic lead and separated from the glass phase; this allows the recovery of approximately 90% of the lead from the glass. However, the resulting glass still contains approximately 1–2 wt% of unrecovered lead, which can be released into the environment. Therefore, in our previous studies [15,18], the reduction-melting process was modified to immobilize or recover the unrecovered lead, and two advanced processes were proposed. First, the reduction-melting was combined with subsequent oxidative melting [15]. In this process, the resulting glass after the reduction-melting was re-melted under an oxidative atmosphere to immobilize the unrecovered lead in the glass. In the oxidation

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stage, the unrecovered metallic lead in the glass was converted to lead oxides incorporated in the SiO₂ network of the glass, and lead immobilization was promoted. The quantity of the lead extracted from the final glass product into water or acid was lower than the Japanese environmental criteria for recycling as construction materials. However, as SiO₂, Al₂O₃, MgO, and NaNO₃ were mixed with the glass in the oxidative melting stage to promote the lead immobilization, it led to an increase in the overall treatment cost. In order to decrease the dosage of these additives, it is necessary to minimize the generation of water-soluble lead compounds in the glass during the reduction-melting.

In contrast to the above-mentioned reductive/oxidative melting, a process that combined the thermal treatment and leaching processes was developed to promote further recovery of the unrecovered lead [18]. In this process, after the reduction-melting, the molten glass was annealed at 700 °C to promote the phase separation of sodium oxides and crystallization of sodium silicates in the glass. As a result, sodium was efficiently extracted in water from the above-mentioned water-soluble sodium compounds [17]. The extracted sodium can be recovered as Na₂CO₃ flux and re-used in the reduction-melting stage. After the water treatment, the residual oxide phase was washed with 1 M HCl to increase the purity of the silica in the oxide phase. In these leaching processes, the lead remaining in the oxide phase was extracted, and silica powder with a purity exceeding 95 wt% was obtained. In order to decrease the lead concentration in the silica powder, it is necessary to promote the generation of water-soluble lead compounds in the glass during the reduction-melting.

Hence, it is important to control the solubility of the unrecovered lead in the glass after reduction-melting in the above-mentioned advanced processes. In our previous study, the factors determining the generation of acid-soluble lead in the melting process were examined [21]; however, the water-soluble lead was not specifically investigated. The purpose of this study is to clarify the water-soluble lead species generated in the reduction-melting of the funnel glass and to identify the factors determining their generation. The funnel glass from a domestic treatment facility for CRT monitors was melted under a reductive atmosphere at different melting conditions, and a water leaching test was performed on the resulting glass. The quantity of the water-soluble lead in the glass was compared with the results of the speciation analysis of the unrecovered lead in the glass.

2. Materials and methods

2.1. Melting experiment

In our previous study, powders of the funnel glass with particle size <1 mm (FG powder) were used for the melting experiment [15]. In the present study, the same powders were used, and their chemical composition is shown in Table 1. The melting experiment

was performed in a manner similar to that of the previous study. In the experiments, a mixture of 20 g FG powder, activated carbon, and Na₂CO₃ was used. The carbon and Na₂CO₃ dosages were varied across the experiments (Table 2). The mixture was put in a 30-mL alumina crucible (Al₂O₃ = 95%), and the crucible was heated in an electric furnace at a specific temperature. The melting temperature and melting time were also varied as shown in Table 2. In the melting process, lead oxides in the funnel glass is reduced, and fine metallic lead particles are generated in the molten glass. The metallic lead particles aggregate, settle, and are separated from the molten glass as an agglomerate. After melting, the crucible was allowed to naturally cool to room temperature. The resulting product was crushed to separate the agglomerate of metallic lead phase from the glass phase. The resulting glass was ground in a mortar and sieved to prepare a 53–149 μm glass powder. This glass powder was evaluated according to the procedure explained in the following section. As shown in Table 2, there were variations in the Na₂CO₃ additions to the FG powder, and the sodium concentrations in the resulting glass varied depending on the Na₂CO₃ additions (Table 1). Based on the sodium concentrations, the resulting glass is classified into two groups, namely sodium-poor glass (N0C3, N1C3, N2C3, N3C3, and N4C3) and sodium-rich glass (N10C0, N10C1, N10C2, N10C3, N10C4, N10C5, N10C3T1300, N10C3T1400, N10C3–2 h, and N10C3–3 h). In the former group, the sodium concentrations were below 20 wt%, whereas in the latter group, the sodium concentrations were over 20 wt%. Detailed information on the FG powders and melting experiments are obtained from the above-mentioned previous study [15].

2.2. Analysis of glass

The chemical composition of the resulting 53–149 μm glass powder was determined by X-ray fluorescence (XRF) analysis using a Rigaku ZSX Primus II system (Rigaku Co., Ltd., Tokyo, Japan). A part of the metallic lead generated by the reduction of the lead oxides remained in the resulting glass. The concentration of such unrecovered metallic lead in the glass was determined by differential scanning calorimetry (DSC) recorded using EXSTAR DSC6000 (Seiko Instruments Co., Ltd., Chiba, Japan). This method was previously employed by Méar et al. [10]. They calculated the metallic lead concentration by comparing the peak area under the endothermic peak corresponding to the melting of metallic lead in a glass material with that under a peak resulting from a reference sample in the DSC analysis. In the present study, approximately 20 mg of each glass powder was used for the DSC analysis, and the same analysis was repeated twice. The average values of the obtained metallic lead concentrations for each glass powder (Table 3(a)) are used later in the discussion.

Table 1
Chemical composition of FG powder and glass after melting experiments (unit:wt%).

Compounds	FG powder	Glass after melting experiment															
		N10C0	N0C3	N1C3	N2C3	N3C3	N4C3	N10C1	N10C2	N10C3	N10C4	N10C5	N10C3T1300	N10C3T1400	N10C3–2h	N10C3–3h	
SiO ₂	45.9	36.7	51.8	54.4	52.8	52.7	51.8	40.0	44.9	44.5	45.4	45.1	50.4	50.0	45.3	45.2	
PbO	25.2	19.0	15.2	10.2	10.4	6.5	5.0	7.4	2.2	2.0	1.2	2.0	0.78	0.97	1.4	1.2	
Na ₂ O	5.8	26.0	7.0	9.9	11.9	16.1	19.0	32.9	32.1	31.2	32.2	32.2	27.1	26.6	32.0	32.0	
K ₂ O	8.9	6.4	10.6	10.1	10.0	9.5	9.1	7.3	7.7	7.7	7.5	6.8	6.7	6.7	7.7	7.6	
CaO	4.0	3.3	4.9	4.7	4.6	4.7	4.7	3.7	3.9	4.5	4.0	4.1	3.9	4.0	4.0	4.0	
MgO	1.5	1.2	1.8	1.9	1.8	1.8	1.8	1.3	1.5	1.5	1.6	1.6	1.6	1.6	1.6	1.5	
Al ₂ O ₃	3.3	3.7	4.1	4.2	4.1	4.1	4.0	3.3	3.8	4.6	4.2	4.4	5.2	5.9	4.1	4.5	
SrO	1.9	1.1	1.5	1.5	1.4	1.5	1.5	1.3	1.3	1.4	1.4	1.3	1.8	1.8	1.4	1.4	
BaO	2.0	1.3	2.0	1.9	1.8	1.9	1.8	1.5	1.5	1.5	1.6	1.6	1.4	1.3	1.5	1.5	
Others	1.5	1.1	1.1	1.3	1.3	1.3	1.2	1.3	1.2	1.1	1.0	0.9	1.1	1.1	1.1	1.1	

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