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Potential and Limitations of Upgrading Waste to Load-bearing Structures**

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Cast Glass Components out of Recycled Glass: Potential and Limitations of Upgrading Waste to Load-bearing Structures

Telesilla Bristogianni ^a, Faidra Oikonomopoulou ^b, Clarissa Justino de Lima ^a, Fred A. Veer ^b, Rob Nijse ^{a,b}

^a TU Delft, Faculty of Civil Engineering and Geosciences, The Netherlands, T.Bristogianni@tudelft.nl

^b TU Delft, Faculty of Architecture and the Built Environment, The Netherlands

Although in theory glass can be endlessly remelted without loss in quality, in practice only a small percentage gets recycled, mainly by the packaging industry. Most of the discarded glass fails to pass the high quality standards of the prevailing glass industry -due to coatings, adhesives, other contaminants or incompatibility of the recipe- and ends up in the landfill. However, employing discarded glass in cast components for building applications can be a way to reintroduce this waste to the supply chain. Such components can tolerate a higher percentage of inclusions, without necessarily compromising their mechanical or aesthetic properties. This paper explores the potential but also the limitations of recycling glass in order to obtain load-bearing components. First, an overview is provided regarding which types of glass reach the recycling plants and the which not, arguing on the reasons behind this selection. Afterwards, a series of experiments is presented, exploring the possibilities of recycling everyday glass waste, from beer bottles and Pyrex® trays to mobile phone screens. Each type of glass waste is initially cast separately to define its flow capability and risk of crystallization. The above information is linked to the X-ray fluorescence (XRF) analyses of the samples prior to recycling. Then, the possibility to mix different glass recipes without fracturing is evaluated. The results point out the types of glass with potential in structural applications, and the overall feasibility of the concept.

Keywords: Glass waste, glass recycling, cast glass components, kiln-casting

1. Introduction

The term glass recycling is almost a synonym to the sole recycling of glass bottles and containers for the purposes of the food packaging industry. The glass packaging industry has invested in the infrastructure for the collection, sorting and processing of glass bottle and container waste, and managed high percentages of waste recovery and recycling, such as a mean of 73,2% in 2015 in the European Union (Eurostat 2016). Glass waste is however a broader term including overall everyday household waste (ex. tableware, ovenware, lighting), building waste (ex. windows, glass tiles), electronic waste (ex. Liquid Crystal Displays and Cathode Ray Tubes), automotive industry waste (ex. laminated windshields) and industrial/laboratory waste to name a few. The percentages of recovery and recycling of such glass waste are rather low, if not zero (ELVIROS 2004). Such glass objects are considered non-recyclable either due to contamination from coatings, adhesives, laminates or even hazardous substances, or due to a required labour intensive demounting process (ex. Window panes, computer screens) (Dyer 2014). The food packaging industry does not accept contaminated glass cullet that will affect the taste of the products, same as the float industry rejects such glass, as it is responsible for the creation of stones and other flaws and reduces the transparency of their glass panes. Since this glass cannot meet the strict criteria to be reused for the same purpose (close-loop recycling), it either ends up in landfills or gets down-cycled (open-loop recycling) into aggregate in concrete, ceramic or pavement products, into abrasive or into foam insulation (Silva et al. 2017; Dyer 2014). What is more, tons of non-contaminated glass waste are simply discarded or down-cycled due to their mismatch in recipe with that of glass packaging. In other words, the lack of facilities and automated processes for separation and handling of different types of glass is- at a great extend- responsible for the rejection of this waste from the close-loop recycling.

Cast glass technology for structural applications can be a strategy for tackling the problem of glass waste rejection due to contamination or glass composition. Cast glass has already been applied in load-bearing applications, such as the self-supporting glass façade of the Crystal Houses in Amsterdam (Oikonomopoulou et al. 2017). On the one hand, building components out of cast glass can tolerate more flaws (bubbles, color shifts etc) than a piece of float glass or a drinking bottle, without compromising the strength or aesthetic quality. On the other hand, the relatively smaller scale of the cast glass factories and often lack of automation leave more room for experimentation. In contrast to the giants of glass processing and their strict specifications, cast glass producers have more freedom in altering the firing schedules and the glass recipes.

Scope of this paper, is the categorization of the glass waste and the understanding of its value as a raw source for the casting of glass building components.

2. Categorization of everyday glass waste

2.1. Prevailing glass families

By clustering the commercial glass waste into families of similar chemical composition, we can set guidelines on how to handle each piece of glass waste and what to expect from the resulting product. In specific, we can define the working and annealing temperature range, the easiness of crystallization, the coefficient of thermal expansion and the possibility of combination with similar glass waste sources. We can also predict the characteristics of the finished products in terms of mechanical, optical and thermal properties.

Everyday glass waste was therefore categorized in the following six families: soda-lime, soda-potash-lime, lead crystal, lead-free crystal, borosilicate and alkali-aluminosilicate glass. Within the soda-lime category- which is also the most prevailing one- three subcategories were set in relation to the manufacturing process of the glass objects, since the production method fine tunes the basic soda-lime recipe. These subcategories are: automated blown, mouth-blown and float glass. Other types of specialty glasses are not included in this research as their contribution to the problem of glass waste is less considerable.



Fig. 1 Main families of commercial glasses.

Figures 2a) and 2b) provide a list of properties for each family of commercial glass. This list will be further used in this research as a guideline for determining the handling temperatures during the casting of the different glass waste.

Properties of various types of commercial glasses				
Glass family	Lead crystal	Lead-free barium strontium		Soda lime (mouth-blown)
Glass name	Schott LF5	CRT colour panel	Corning 9068 (Colour TV panel)	Cristalica, Spruce Pir
Young's modulus E in kN/mm ²	59			
Thermal expansion coefficient α (20°C;300°C) in 10 ⁻⁶ K ⁻¹	10,6	9,7-10,9	9,9	
Density in g/cm ³	3,22	2,75-2,79	2,69	
Forming temperature (viscosity=log4 in dPas) in °C				95
Forming temperature (viscosity= log5 in dPas) in °C				85
Forming temperature (viscosity=log6 in dPas) in °C				75
Softening point (viscosity=log7,6 in dPas) in °C	585		688	65
Annealing point (viscosity=log13 in dPas) in °C	411	501	503	55
Strain point (viscosity=log14,5 in dPas) in °C			462	45
Sources	(SCHOTT 2014b)	(Mear et al. 2006) (Shelby 2005)	(Campbell et al 1990) (Seward III et al. 2001)	(Spruce Pine Batc

Fig. 2a) Properties of commercial glasses.

Soda-lime silica glass													
Blown, automated: Glass Bottles						Blown, automated: Drinking glass			Mouth-blown glass				
Wine bottle mix						Champagne glass			Spruce Pine transparent blue				
1. Clear		2. Light blue		3. Light green		4. Green							
Compound name	Content (wt%)	Compound name	Content (wt%)	Compound name	Content (wt%)	Compound name	Content (wt%)	Compound name	Content (wt%)	Compound name	Content (wt%)	Compound name	Content (wt%)
SiO2	73.356	SiO2	72.628	SiO2	73.358	SiO2	71.124	SiO2	73.241	SiO2	71.768		
Na2O	11.882	Na2O	11.761	CaO	11.888	Na2O	12.903	Na2O	12.682	Na2O	14.288		
CaO	9.878	CaO	9.746	Na2O	11.477	CaO	11.072	CaO	10.883	CaO	6.834		
MgO	2.165	MgO	3.129	Al2O3	1.305	Al2O3	2.875	Al2O3	1.621	Al2O3	2.306		
Al2O3	1.261	Al2O3	1.603	MgO	1.013	K2O	0.882	MgO	1.277	CuO	2.038		
K2O	0.639	K2O	0.661	K2O	0.397	Fe2O3	0.446	S	0.149	Fe2O3	0.835		
SrO	0.203	S	0.19	Fe2O3	0.257	MgO	0.239	TiO2	0.052	ZnO	0.759		
BaO	0.184	Fe2O3	0.144	TiO2	0.073	Cr2O3	0.211	Cl	0.027	K2O	0.38		
S	0.143	TiO2	0.049	Cr2O3	0.062	TiO2	0.07	K2O	0.025	BaO	0.355		
ZrO2	0.085	Cl	0.026	BaO	0.047	P2O5	0.045	P2O5	0.022	Sb2O3	0.161		
Fe2O3	0.069	P2O5	0.022	S	0.025	Cl	0.044	ZrO2	0.013	MgO	0.133		
TiO2	0.045	ZrO2	0.017	SrO	0.021	S	0.038	SrO	0.008	ZrO2	0.046		
Cl	0.031	ZnO	0.009	ZrO2	0.02	BaO	0.025			Co3O4	0.04		
ZnO	0.024	PbO	0.008	Cl	0.018	SrO	0.017			S	0.04		
P2O5	0.021	SrO	0.006	PbO	0.016	ZrO2	0.009			SrO	0.012		
PbO	0.013			ZnO	0.011					P2O5	0.005		
				P2O5	0.01								
				Rb2O	0.003								

Soda-lime silica glass									
Mouth-blown glass		Float glass							
Spruce Pine transparent orange		PPG Starphire (extra clear)		PPG clear		PPG furnace waste aquamarine		PPG furnace waste light green	
Compound name	Content (wt%)	Compound name	Content (wt%)	Compound name	Content (wt%)	Compound name	Content (wt%)	Compound name	Content (wt%)
SiO2	73.144	SiO2	74.563	SiO2	74.214	SiO2	73.108	SiO2	67.41
Na2O	15.393	Na2O	13.323	Na2O	12.438	Na2O	14.346	Na2O	13.676
CaO	7.093	CaO	8.905	CaO	10.029	CaO	7.888	Al2O3	8.828
Al2O3	2.058	MgO	3.006	MgO	2.859	MgO	3.947	CaO	5.943
ZnO	0.886	S	0.105	Al2O3	0.142	Al2O3	0.311	ZrO2	2.975
BaO	0.411	Al2O3	0.035	Fe2O3	0.124	S	0.159	MgO	0.431
K2O	0.411	Fe2O3	0.015	S	0.084	Fe2O3	0.087	TiO2	0.213
MgO	0.258	Cl	0.014	K2O	0.043	K2O	0.078	Fe2O3	0.171
Sb2O3	0.223	K2O	0.012	P2O5	0.038	TiO2	0.035	S	0.101
Fe2O3	0.027	P2O5	0.01	Cl	0.024	ZrO2	0.014	SrO	0.089
ZrO2	0.024	ZrO2	0.007	SrO	0.005	Cl	0.013	K2O	0.073
S	0.023	SrO	0.005			P2O5	0.009	HfO2	0.055
PbO	0.021					SrO	0.005	P2O5	0.02
SrO	0.012							Cl	0.015
Cl	0.01								
P2O5	0.005								

Potash-soda-lime glass (optical glass)			
Schott B270 lenses		Czechoslovakia bowl	
Compound name	Content (wt%)	Compound name	Content (wt%)
SiO2	71.802	SiO2	82.632
Na2O	10.138	Na2O	6.488
K2O	6.275	CaO	4.146
CaO	5.168	K2O	3.2
ZnO	2.198	MgO	2.755
Al2O3	2.083	SO3	0.281
TiO2	1.765	Al2O3	0.219
Sb2O3	0.403	Cl	0.1
MgO	0.041	TiO2	0.06
BaO	0.03	Fe2O3	0.045
Cl	0.022	PbO	0.028
S	0.018	P2O5	0.023
P2O5	0.017	ZnO	0.01
Fe2O3	0.016	ZrO2	0.009
ZrO2	0.008	Rb2O	0.004
SrO	0.006	Br	0.002
Rb2O	0.005		
PbO	0.005		

Lead Crystal					
Schott LF5 lead crystal		G210		Crystal D'Arques 24% Lead Bowl	
Compound name	Content (wt%)	Compound name	Content (wt%)	Compound name	Content (wt%)
SiO2	53.731	PbO	47.002	SiO2	63.229
PbO	36.643	SiO2	44.714	PbO	24.799
K2O	5.387	K2O	3.936	K2O	7.753
Na2O	3.771	Na2O	3.296	Na2O	2.842
Al2O3	0.198	S	0.231	CaO	1.2
SO3	0.134	Sb2O3	0.215	S	0.075
CaO	0.114	CaO	0.212	Al2O3	0.051
NiO	0.018	Al2O3	0.203	ZrO2	0.026
P2O5	0.005	Cl	0.086	Fe2O3	0.024
		MgO	0.06		
		Fe2O3	0.046		

Alkali-barium silicate glass	
CRT computer screen (panel)	
Compound name	Content (wt%)
SiO2	61.505
SrO	8.056
BaO	8.039
Na2O	7.21
K2O	6.776
ZrO2	3.587
Al2O3	2.304
CaO	1.109
TiO2	0.378
Sb2O3	0.343
MgO	0.296
SO3	0.153
Fe2O3	0.095
CuO	0.055
Cl	0.028
ZnO	0.027
P2O5	0.023
NiO	0.018

Borosilicate glass			
Schott lab glassware			
Compound name	Content (wt%)	Compound name	Content (wt%)
Boron not traced therefore Si content appears high			
SiO2	93.024	SiO2	80
Na2O	3.548	B2O3	13
Al2O3	2.725		
K2O	0.513		
Cl	0.051		
ZrO2	0.049		
Fe2O3	0.036		
TiO2	0.033		
P2O5	0.012		
ZnO	0.005		
SrO	0.002		
Rb2O	0.002		

Alkali-aluminosilicate glass	
Mobile phone screen thin glass	
Compound name	Content (wt%)
SiO2	63.062
Al2O3	12.958
K2O	11.12
MgO	6.235
Na2O	5.603
ZrO2	0.843
CaO	0.062
S	0.039
Fe2O3	0.027
Cl	0.017
TiO2	0.014
HfO2	0.013
P2O5	0.006
SrO	0.002

Fig. 3 Composition of selected waste glass by category (XRF analysis).

3. Recycling experiments and interpretation of the results

3.1. Experimental set-up

In this section, an explanation of the set-up parameters is provided. Regarding the casting method for the sample production, the two techniques listed below were followed, according to the maximum operating temperature needed:

- a) Kiln-casting employing investment silica-plaster moulds. For this method, one kiln is used for the melting and annealing of the glass and therefore, the feeding of the moulds with glass takes place inside the kiln. Three different types of moulds were produced:
 - Crystalcast M248, powder to water volume ratio 2,75:1. The product consists of cristobalite, quartz and gypsum (Gold Star). For firings above 1000°C, the crystalcast moulds are reinforced by an exterior layer of heat-resistant concrete.
 - Ransom & Randolph (R&R) Glass Cast 910, powder to water mass ratio 10:2,8. The product consists of cristobalite, quartz, mullite, calcium sulfate and fibrous glass (Ransom & Randolph).
 - Heat-resistant concrete coated with a 1mm thick layer of Mold Mix 6 by Zincar, which is a high alumina putty coating with glass reinforcement fibers (ZRCI 2017). Two layers of EKamold® spray are applied as top coating. This product is an ethanolic coating based on hexagonal boron nitride (ESK 2013). This mould was used for a firing at 1250°C.

The glass was introduced in the moulds either directly (“free-set”) or indirectly by being placed in terracotta flowerpots that were positioned above the moulds. The heating ramp was set at 50°C/hr. Regarding the quenching applied to prevent the crystallization of the components, the samples are either manually quenched below their softening point, by opening and closing the kiln door in repetition or mechanically by setting the kiln controller at the “As Fast As Possible” (AFAP) function. The later cooling process requires more time versus the manual, abrupt quenching.

- b) Melt-quenching technique employing high-alumina crucibles and steel moulds. This method is preferred in this research for castings above 1250°C, due to the high thermal and chemical resistance of the alumina crucibles. For this method, two kilns are needed for the melting and annealing of the glass respectively. The glass is molten in Coors™ high-alumina crucibles and poured at atmospheric conditions in steel moulds that are preheated at 500°C. Upon quenching, the samples are placed in the annealing oven together with the steel mould. The heating rate used was 17,5C°/min.

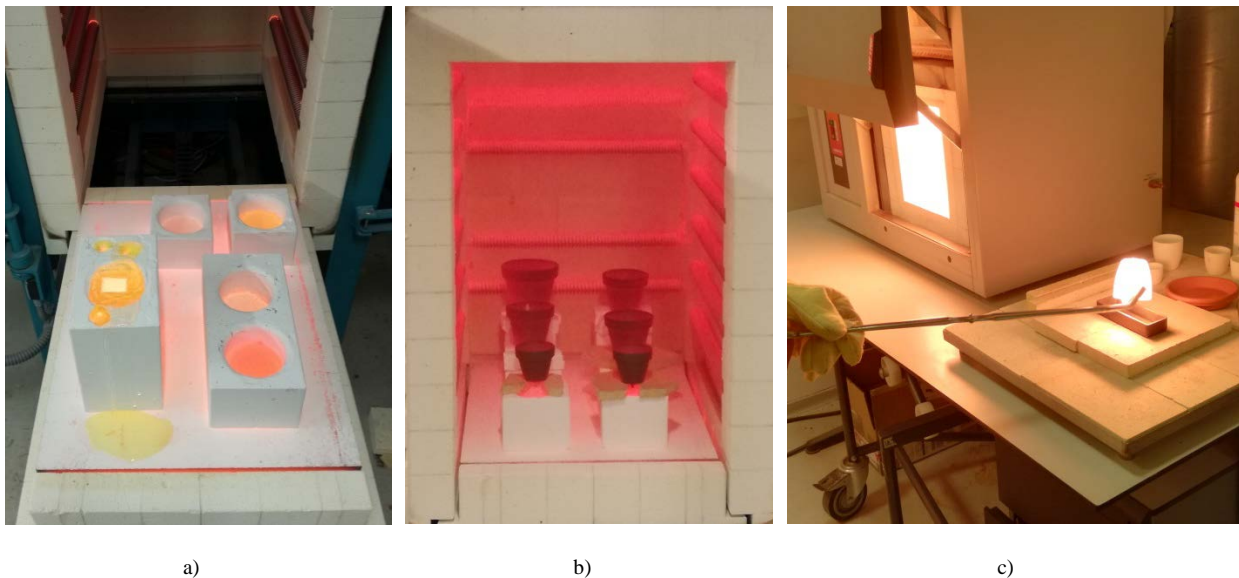




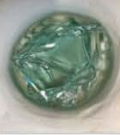




Fig. 4a) Kiln-casting in Crystalcast moulds with glass directly placed in the moulds (free-set), b) Kiln-casting in Crystalcast moulds employing the flowerpot technique, and c) Hot pouring in steel mould.

An overview of the casting experiments including the casting set-up and the results is provided in the subsequent pages.

		Casting temperature in °C											
		860	940	950	970	1200	1250	1350	1450	1500			
Soda-lime glass (blown)	Before firing												
	Data	Spruce Pine transparent orange	R&R Glass-Cast 910 Kiln-casting Flowerpot method Kiln-operated No (different hues of orange used) Full No No										
Soda-lime glass (float)	Before firing												
	Data	PPG Starphire (extra clear)	R&R Glass-Cast 910 Kiln-casting Flowerpot method Kiln-operated No Full No No										
Soda-lime glass (float)	Before firing												
	Data	PPG clear											
Soda-lime glass (float-furnace waste)	Before firing												
	Data	PPG furnace waste aquamarine	R&R Glass-Cast 910 Kiln-casting Flowerpot method Kiln-operated No Yes Yes No										








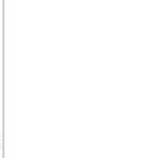

Crystalcast M248
Kiln-casting
Free-set on mould
Abrupt: door open
Yes
Top surface only
No
Yes










Crystalcast M248
Kiln-casting
Free-set on mould
Abrupt: door open
Yes
Top surface only
No
Yes



Crystalcast M248
Kiln-casting
Free-set on mould
Abrupt: door open
Yes
Top surface only
Yes
Yes

		Casting temperature in °C										
		860	940	950	970	1200	1250	1350	1450	1500		
Glass type	Data	Before firing										
												
Soda-lime glass (bottle glass)	Clear bottle (Spa)											
	Mould type	Crystalcast M248										
	Casting method	Kiln-casting										
	Glass feeding	Free-set on mould										
	Quenching	Abrupt: door open										
	Homogeneous melt	Yes										
Crystallisation	Top surface											
Top surface creasing	No											
Transparency	Yes											
Soda-lime glass (bottle glass)	Green beer bottle glass (Stella; 860°C; Heineken; 970°C; 1200 °C)											
	Mould type											
	Casting method	Terracotta (sample could barely flow down to the investment mould)										
	Glass feeding	Kiln-casting										
	Quenching	Flowerpot method										
	Homogeneous melt	Kiln-operated										
Crystallisation	No											
Top surface creasing	Full											
Transparency	-											
Soda-lime glass (bottle glass)	Light green wine Bottle (from USA)											
	Mould type											
	Casting method	Terracotta (sample did not flow down to the investment mould)										
	Glass feeding	Kiln-casting										
	Quenching	Flowerpot method										
	Homogeneous melt	Kiln-operated										
Crystallisation	No											
Top surface creasing	Full											
Transparency	-											
Soda-lime glass (bottle glass)	Mould type											
	Casting method	Terracotta (sample did not flow down to the investment mould)										
	Glass feeding	Kiln-casting										
	Quenching	Flowerpot method										
	Homogeneous melt	Kiln-operated										
	Crystallisation	No										
Top surface creasing	Full											
Transparency	-											

		Casting temperature in °C										
		860	940	950	970	1200	1250	1350	1450	1500		
Aluminosilicate glass	Data	Before firing										
	Mobile phone screen thin glass											
	Mould type											
	Casting method											
	Glass feeding											
	Quenching											
	Homogeneous melt											
Crystallisation												
Top surface creasing												
Transparency												
Combination: Lead crystal and float glass	Data											
	33% Crystal D'Arques (bottom) + 67% ppG starphire (top) per mass											
	Mould type											
	Casting method											
	Glass feeding											
	Quenching											
	Homogeneous melt											
Crystallisation												
Top surface creasing												
Transparency												
Combination: Borosilicate and float glass	Data											
	32% Schott lab glassware DURAN (bottom) + 68% ppG starphire per mass (top)											
	Mould type											
	Casting method											
	Glass feeding											
	Quenching											
	Homogeneous melt											
Crystallisation												
Top surface creasing												
Transparency												
Aluminosilicate glass	Data											
	Crystalcast M248											
	Kiln-casting											
	Free-set on mould											
	Abrupt: door open											
	Reaction with mould											
	Porous structure											
Transparency												
Aluminosilicate glass	Data											
	Crystalcast M248											
	Kiln-casting											
	Free-set on mould (Glass decayed the mould)											
	Abrupt: door open											
	Seems so, however the glass cracked.											
	Transparency											
Aluminosilicate glass	Data											
	Crystalcast M248											
	Kiln-casting											
	Free-set on mould											
	Abrupt: door open											
	Seems so, however the glass cracked in small pieces											
	Transparency											
Aluminosilicate glass	Data											
	Stainless steel											
	Hot poured (molten in crucible)											
	Coors high-alumina											
	In atm. conditions											
	Yes											
	No											
Transparency												

3.2. Casting of individual samples

The previous pages present the results of the castings of samples from the six selected glass families at different temperatures. The samples are initially evaluated on their workability. As the collection of samples includes glasses that have been developed for other production techniques (automated-blowing, float line, automated draw etc), implications are expected when attempting to cast them, especially with the slower method of kiln-casting. Ideally, the glass samples should be able to flow and homogenize at temperatures below 1000°C, both to increase the energy savings but also to simplify the prerequisites of their mould. Aim of the research is thus to identify the glasses that have lower working temperatures and resistance to crystallization from the glasses that are difficult to cast and need extra attention. For this identification step, each glass is cast individually. The following observations and conclusions can be gathered for each glass family:

- Soda lime/ blown, automated (Container glass such as beer/wine/soda bottles, drinking glasses):
These glasses are developed for the automated blowing process, therefore they need to be stiff enough to keep their shape once blown in a mould. Although this prerequisite decreases the workability of the glass, this fact is counteracted by the mechanically applied air pressure for the moulding. This attribute was prevailing during the casting of these glasses, since the samples needed temperatures higher than 1000°C in order to flow and homogenize. Moreover, all samples were very susceptible to crystallization. Abrupt quenching was necessary to avoid full crystallization and to confine the problem only to the top surface. More specifically, samples cast at 860°C would not flow but only partially fuse, and would fully crystallize with mechanical quenching. At 950°C the drinking glass sample failed to homogenize and completely crystallized despite its fast cooling. Likewise, the samples cast at 970°C and mechanically quenched were fully crystallized and incomplete, as the glass could not easily flow from the flowerpot down to the mould. Only when cast at 1200°C and abruptly cooled down to 600°C were the resulting samples transparent. This transparency was evident after the top crystallized surface¹ and the bottom surface in contact with the investment mould were polished.

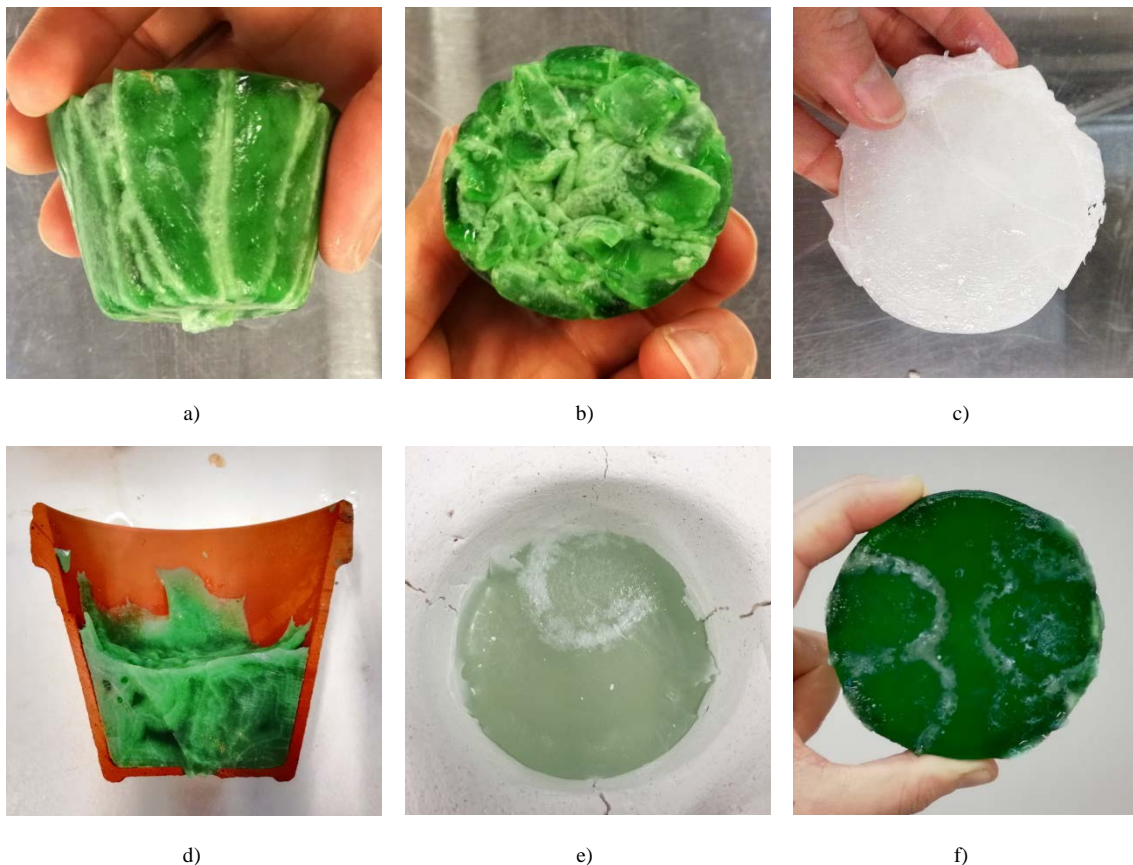


Fig. 5a), b) Beer bottle starting to fuse inside the flowerpot, at 860°C, c) drinking glass completely crystallized at 950°C, d) beer bottle fused inside the flowerpot and partially crystallized, e) clear bottle cast at 1.200°C with top surface crystallization linked to the contact of the glass to the mould, f) beer bottle cast at 1.200°C.

¹ This superficial layer of crystallization seemed to be linked with contamination from the walls of the investment mould, while the glass was flowing downwards.

From the XRF data obtained, we can observe that all above tested glasses have a similar composition. Mainly we see a range of 71,12-73,99 per weight percentage (wt%) of SiO_2 , 10,95-12,90 wt% Na_2O , and 9,74-11,88 wt% CaO . Calcium oxide (CaO , lime) is increasing the softening point and the sagging temperature, thus the observed high viscosity of the samples (Zschimmer 2013). Although this is required for the automated blowing process, the increased softening point impediments the casting. The high percentage of lime is also responsible for the susceptibility to crystallization. According to Zschimmer, lime percentages above 10% promote devitrification. This is to be taken into account when working with this glass sub-family.

It should be also mentioned that although the tested glasses have almost the same composition and viscosity, their colour is affecting their setting time. Kitaigorodskii et al. (1934) and Burch et al. (1938) proved that glasses with identical basic composition and viscosity characteristics, differ regarding their working range. In specific, dark coloured glasses, due to their greater heat loss by radiation rate, tend to set much faster than the equivalent transparent or light coloured samples. Holscher et al. (1943) suggest chromium emerald green glasses of basic composition SiO_2 74,0 wt%, Na_2O 16,0 wt%, CaO 10,0 wt%, Fe_2O_3 0,035 wt% and Cr_2O_3 0,25 wt% to be one of the most rapid cooling systems after dark green Fe-Mn systems. In our tested samples that could be easily verified by the intense luminosity of these samples at 1200°C in comparison to the transparent ones. The fast setting of darker colours should be considered when the hot-pouring method is employed.

Despite the challenges to cast this sub-family, glass container waste is still the most prevailing one. In that sense, it is worth exploring the derailing of sorted- yet discarded by the packaging industry -glass waste, from the landfill to the building sector. As an advantage, the iron in the green and amber (combined with Sulfur) glasses can provide excellent UV-radiation protection (Shelby 2005) and are thus worth to be considered in facades.

- Soda lime/ mouth-blown (artifacts from the glass blowing studio at Southern Illinois University): Glass studios, either commercial or academic, produce quite some glass waste during their trial and error explorations. Especially the discarded pieces by the glassblowing hotshop containing colour are not reused for blown objects, as this would imply the contamination of the clear transparent batch in the furnace.

In contrast to the above glass category, these glasses are meant to be processed according to the power of the human lung, therefore they cannot be as stiff. Also, the artists require a prolonged working time to process their piece, thus a glass that will not set as fast as the container glass. Correspondingly, the content of lime (CaO) in this composition is found around 6,8-7,1 wt% and of sodium oxide (Na_2O) at 14,3-15,4 wt%. The glass samples thus could slowly flow down from the flowerpots to the moulds at 860°C, a temperature considerably lower than the one necessary for the machine-blown glass objects of the same glass family. However, the lack of abrupt quenching can still induce full crystallization at the samples. The differences in the working range between the colour variations are valid also here. Overall though, these coloured glasses are usually engineered to be compatible and in that sense beautiful colour patterns can emerge from their combination. Crystallized samples can lead as well to interesting marble-like smooth components. Since the mechanical properties of these objects are expected to be improved during the devitrification, experimental testing of their strength should follow this research, in order to explore their value as building components.



Fig. 6 a) Set-up of the kiln, b), c) glass ceramic samples resulting from the mouth-blown glass waste.

Cast Glass Components out of Recycled Glass: Potential and Limitations of Upgrading Waste to Load-bearing Structures

- Soda lime/ float (window glass, waste glass from furnace clean-up):
The float glass samples present a bit higher weight percentages of SiO_2 74,21-74,56 and Na_2O 12,44-13,32, and lower CaO percentage of 8,91-10,03 in comparison to the automated-blown glass. Magnesium Oxide (MgO) levels are also higher by 1-1,5% wt%. The XRF analysis was conducted at only one surface of the samples which is assumed to be the top surface since no contamination by the tin bed is observed. The glass would be very viscous at 860°C , flowing very slowly. Full crystallization occurred by mechanical quenching at the extra clear sample (PPG Starphire). These glasses could be cast homogeneously at 1200°C and presented only surface crystallization with abrupt cooling. Here the difference in the crystallization pattern between the clear and the extra clear float samples should be mentioned. The clear sample presented a subtle translucent finishing at the top surface and a couple of local areas of mild crystal formations. The extra clear (PPG Starphire) glass however, showed a more intense crystallization pattern at the top surface, with visible crystal clusters.

It is not clear- when comparing the two glass compositions- why the extra clear sample is more prone to crystallization, but this could also be related to a possible contamination of the top surface from the tin bath. An XRF analysis of the cast sample should be conducted to further investigate this. To better define the extra clear glass, a comprehensive glass thermal analysis (Differential Scanning Calorimetry DSC) was performed employing a STA 449 F3 Jupiter[®] apparatus. The test showed a stable glass, but with a crystallization peak at around 740°C . The glass transition temperature was found at the region of 570°C . With proper cooling- fast enough to avoid the crystallization at the above mentioned peak- this glass can result into very clear and transparent castings.



a)

b)

c)

Fig. 7 a) PPG Starphire kiln-cast at 860°C and mechanically quenched, b) PPG Starphire sample kiln-cast at 1.200°C with top surface crystallization, c) PPG clear float sample kiln-cast at 1.200°C .

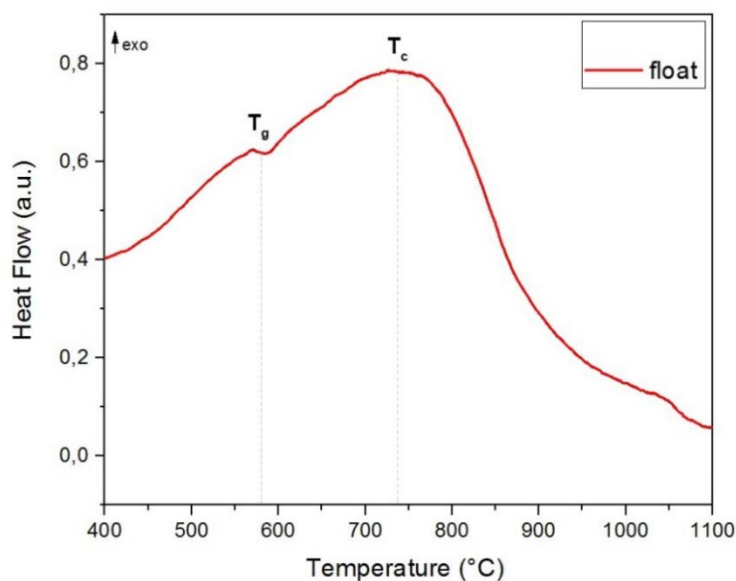


Fig. 8 DSC analysis pointing out the glass transition temperature and crystallization peak temperature of PPG Starphire.

Regarding the waste from the float production furnace clean-up, this refers to glass that may be sitting at the bottom of the furnace for years. The glass slowly decays the refractory materials at the base and absorbs part of their elements in its composition. The resulting glass- although in rough lines similar in composition to the original soda-lime one, can be quite unpredictable in its exact composition and therefore in its casting behavior. For example, the “PPG light green waste glass” was a very viscous glass (could not flow at 860°C) but also extremely resistant to crystallization. The increased viscosity can be explained by the high content of alumina (8,8 wt%). According to Zschimmer (2013), although alumina reduces the melting point of soda-lime-silica glasses when introduced in small amounts, in weight percentages of more than 7% it has the opposite effect. The increased alumina content in combination with the relatively low percentage of lime (5,94 wt%) is what makes this glass so resistant to crystallization. It is also interesting to point out the content of 2,98 wt% of zirconia (ZrO_2) in combination with traces of hafnia (HfO_2). Zirconia is a refractory material found as paving on the furnace bottom (Clark-Monks et al. 1980), especially in cases where very corrosive glasses are melted (Bray et al. 2001). Moreover, Alumina-Zirconia-Silica (AZS) refractories are often used by the glass industry in favour of high-alumina refractories, which tend to more easily corrode and release alumina in the molten glass (Bray et al. 2001). The usage of such refractory products can explain the high content of this glass in alumina and zirconia. Zirconium minerals contain hafnium, in a range of 1,5-2,5% Hf/Zr+Hf or more (Nielsen 2000), justifying the traces of hafnia in this glass. Karell et al. (2007) mention that zirconia increases the viscosity of the melt- as observed- but is also used as a substitute to PbO in lead-free crystal glasses, as it increases the refractive index of the glass and thus the light dispersion. The later information can be linked with the high optical quality of the cast sample.



Fig. 9 a), b) PPG furnace waste green glass before and after casting at 860°C, c), d) PPG furnace waste aquamarine cast at 860°C and 1.200°C respectively.

Cast Glass Components out of Recycled Glass: Potential and Limitations of Upgrading Waste to Load-bearing Structures

The “PPG furnace waste aquamarine” sample did not exhibit serious contamination from the refractory materials and had a composition very close to that of standard float glass. At 860°C the sample was viscous but could only flow slowly. By inducing mechanical cooling, partial crystallization occurred, with some glassy amorphous regions still preserved within its mass. The same glass could be cast homogeneously at 1200°C and presented minor surface crystallization as did sample “PPG clear”.

Despite the unpredictable character of this type of glass waste, interesting colours and patterns can emerge from the casting of these- enriched through their prolonged contact with the furnace- glasses.

- Soda-potash lime (optical lenses, tableware):

Potassium oxide (K_2O) is often added in soda-lime-silica systems to achieve extra white clear glass (Zschimmer 2013). K_2O lowers the melting point of the glass yet increases the thermal expansion coefficient. Such alkali-lime silica systems containing considerable amounts of soda and potash and reduced amounts of lime are preferred for hand- pressing, since these glasses are soft and easy to adapt to the shape of the steel mould while still viscous (Rosenhain 1908). In this category, the optical glass B270 by Schott is used as a reference, since the authors have continuously experimented with this glass due to its good optical qualities and workability at 950°C (Bristogianni et al. 2017). A complete replacement of soda by potassium oxide is found in the pressed historical Bohemian crystal artifacts (Rosenhain 1908). Contemporary Czech (or formerly Czechoslovakian) glass pressed objects however- like the one tested in the scope of this research, may contain considerably less percentages of K_2O , and more soda and silica. The glass obtained after firing at 950°C was transparent and extra clear, yet presented intense creasing at the top surface. Possibly, a small increase in temperature would result in a better quality casting.



Fig. 10 a) and b) Czech glass cast at 950°C. Post-processing is required to remove the shrinkage of the top surface and reveal the actual transparent glass

- Lead crystal (tableware):

Lead silica glass was a glass type commonly used for high quality table and ornamental ware before the use of lead (II) oxide (PbO) was restricted due to toxicity. This composition was preferred by the manufacturers because of the high refractiveness that the PbO would attribute to the glass, adding brilliancy to the glass artifacts (Rosenhain 1908). Shelby (2005) explains that due to the relatively weakness of the $Pb-O$ bonds caused by the low field strength of the large Pb^{2+} ions, the lead-silicate network can be easily disrupted. This justifies the low glass transformation temperature of this glasses. As these glasses are relatively soft, they are selected by the manufacturers, when complicated manipulations during production are required (Rosenhain 1908). Lead glasses also have X-ray protective properties, which escalate with the increase of lead content, and are independent from the radiation quality (Singer 1936). Yet, PbO lowers the Young modulus and the hardness of the glass and significantly increases its density (Zschimmer 2013), factors that should be seriously considered when evaluating the use of such glasses for structural applications.

In this category, Schott LF5 (≈ 36 wt% PbO) and Gaffer G210 (≈ 44 wt% PbO) lead crystal are used as a reference in the XRF analyses, as the authors have previously conducted successful castings at 860°C, achieving extra clear glass without crystallization upon mechanical cooling. The tested sample was a pressed

crystal bowl of lower lead content (≈ 24 wt%), which is a typical percentage for such tableware. The glass was successfully cast at 950°C to a clear glass without crystallization (upon abrupt quenching). At this temperature, the sample is expected to have a much lower viscosity in comparison to soda-lime glass. Yet it is interesting to mention that patterns from the initial design of the bowl were preserved at the bottom of the sample in combination with creasing at the top surface. More experiments are required to determine the flowability of this glass at this temperature, but nonetheless this glass can result to very transparent castings.



Fig. 11 a) and b) Pressed crystal glass before and after casting at 950°C . The pattern of the glass remained as a trace at the bottom surface of the sample. Upon post-processing, the glass will be transparent.

- Lead-free crystal (CRT screen/panel):
Due to the health and environmental concerns linked with the toxicity of lead and other heavy metals, various protocols have been issued around the world with the aim to reduce its use. Regarding glass manufacturers, they would chose PbO either to increase the refractive index adding brilliance to the glass, or to provide radiation protection in nuclear plants, hospitals, TV-tubes etc. Currently, lead is replaced with barium, strontium and zirconium, although some of the alternative elements (barium in particular) can be quite toxic themselves, only less than lead (Scoullou et al. 2001). The resulting glasses present good optical quality, lower density than lead crystal, and x-ray shielding capacity which is however reduced and dependent on the X-ray quality (Singer 1936).

In this section, the casting of the panel of a colour display Cathode Ray Tube (CRT) is attempted. CRT tubes have been used in TV and computer screens before the emergence and dominance of Liquid-Crystal Display (LCD) technology. Although currently scarcely in use, CRT glass waste constitutes an accountable percentage of the municipal waste in the European Union (Hreglich et al. 2001; Bernardo et al. 2005). Since CRTs are not in production anymore, the route of their recycling into the same product has closed, leaving a question mark on how and where this glass can be used (Edgar et al. 2008). Nonetheless, the glass formulation for the CRTs is of fine quality, employing- among others- expensive barium (Ba) and strontium (Sr) oxides (Compton 2003; Bernardo et al. 2005). More specifically, a typical CRT tube consists of three parts: the faceplate (panel), funnel, and neck tubing. The panel, in specific, is a lead-free barium –strontium glass that protects the viewers from the harmful X-rays (Compton 2003).

A sample of a CRT panel was successfully cast at 950°C , resulting in a clear, homogeneous glass of grey hue that did not crystallize upon abrupt quenching. Indeed, this glass composition, rich in Ba and Sr, is expected to be very resistant against crystallization (Kosmal et al. 2017). Of particular interest is however the white dotted pattern that was observed at the sample's top surface. The white substance- possibly surface crystallization induced by contaminants in the air circulated inside the kiln- was easily removed by submerging the sample into water.

Cast Glass Components out of Recycled Glass: Potential and Limitations of Upgrading Waste to Load-bearing Structures

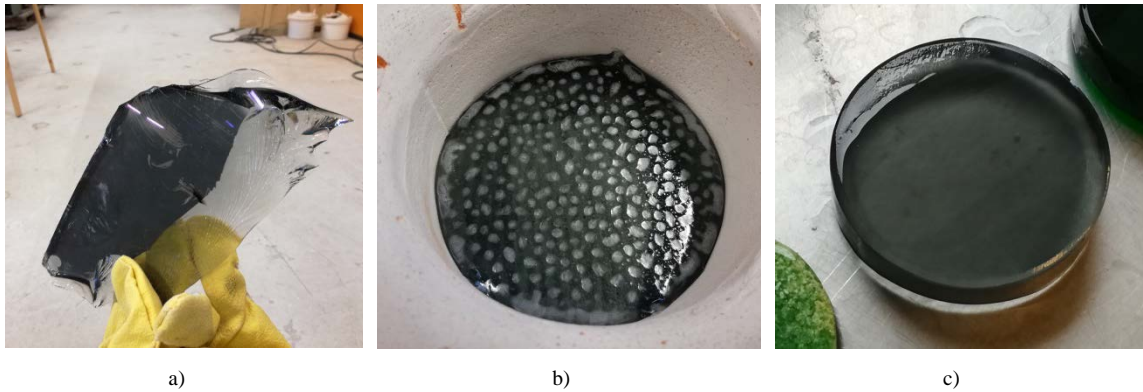


Fig. 12 a) CRT panel as retrieved from a computer screen, b) Top surface crystallization of the sample cast at 950°C, c) The sample resulted into a transparent grey glass after being post-processed

A DSC analysis was conducted employing a STA 449 F3 Jupiter[®] apparatus. The test showed a stable glass with a shallow crystallization peak at around 700°C. Its glass transition temperature was found at the region of 550°C-580°C. The stability of the glass and its high visual resulting quality prove the potential of this glass for casting glass building components.

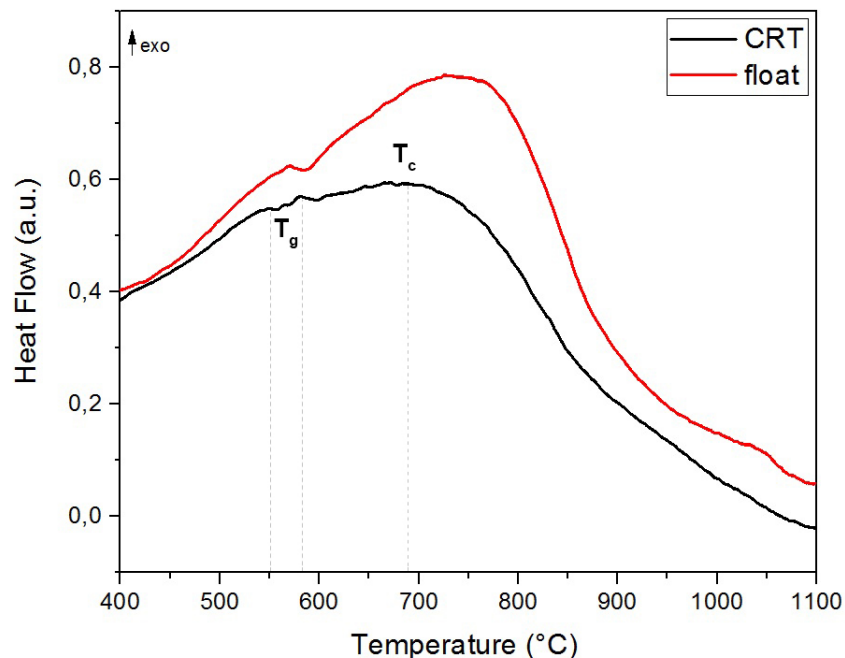


Fig. 13 DSC analysis pointing out the glass transition temperature and crystallization peak temperature of the CRT sample. The temperature to heat flow curve of PPG Starphire float glass is included in the diagram as a point of reference.

- Borosilicate (laboratory tubes):

Borosilicate glass can be attractive for structural applications at demanding environmental conditions, due to its good optical properties and its high thermal and chemical durability (Schott 2014b). It is commonly found in thermal shock resistant cookware and laboratory equipment. Its higher working temperature in comparison to that of soda-lime adds implications to its processing. Guidelines regarding recycling commonly advise not to discard borosilicate objects together with container glass. Unless chemically contaminated and thus hazardous, this good quality glass often ends up in landfills. Investigations for its downcycling into micro-filler for concrete (Korjamins et al. 2012), glass foams (Chinnam et al. 2014) and other ceramic material applications are currently conducted.

A piece of DURAN[®] labware by Schott was cast at 1.200°C and resulted into a clear homogeneous glass after being abruptly cooled. The top surface was completely flat and showed some local crystallization. As this is a very favourable glass, next research steps will test its flowability at the same temperature with the goal to kiln-cast it into reinforced investment moulds via the flowerpot technique.

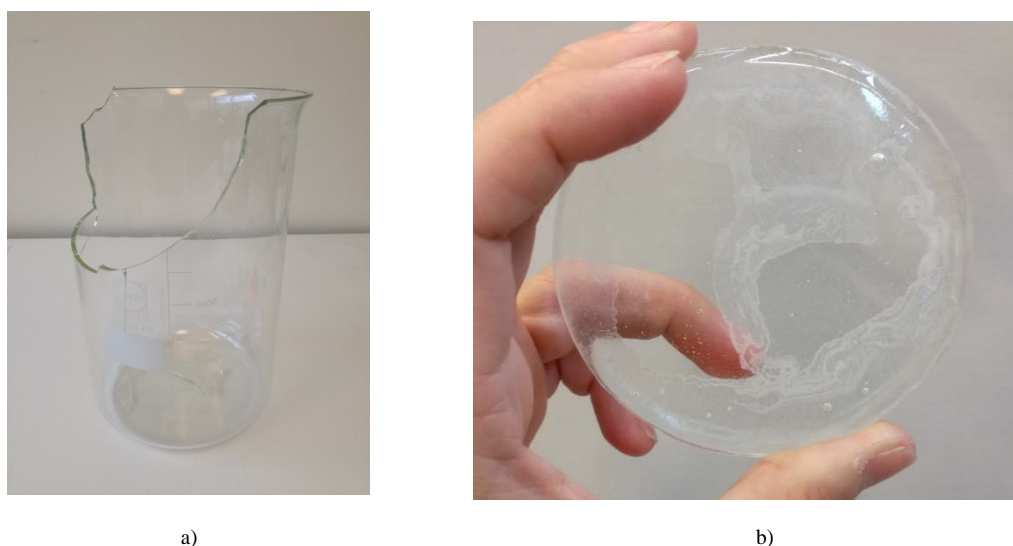


Fig. 14 a), b) Borosilicate labtube before and after casting at 1.200°C.

- Alkali-aluminosilicate (mobile phone screen): Alkali-aluminosilicate glasses are characterized by high glass transition temperatures and excellent mechanical properties such as increased hardness, and scratch and sharp contact damage resistance (Corning 2017; Schott 2014b). Their high alkali content (>10%) enables the ion exchange with bigger alkali ions (ex. potassium bath) that results in a considerably improved surface compressive strength (Schott 2014b). This glass can be drawn via an automated process into very thin sheets of glass (0,4-2mm) that find applications in the screens of smartphones, laptops, tablets and other similar devices (Corning 2015). Due to its outstanding mechanical properties and the fact that touchscreens of that kind are an upcoming source of waste, the recycling of alkali-aluminosilicate glass into building components becomes intriguing. Yet, their extremely high working temperatures render the material challenging for hot-pouring and kiln-casting.

An initial test was conducted by melting a mobile phone screen glass in a high-alumina crucible. At 1.500°C the sample still presented a very high viscosity, but some glass drops managed to flow and resulted in a glass of high air-bubble content. The results of the kiln-casting test at 1.250°C were quite unexpected as the glass partially corroded the silica-plaster mould and formed- probably as a reaction to the mould- a three-dimensional sponge structure of opaque white colour. Further testing is required to understand the reasons behind this foam formation.

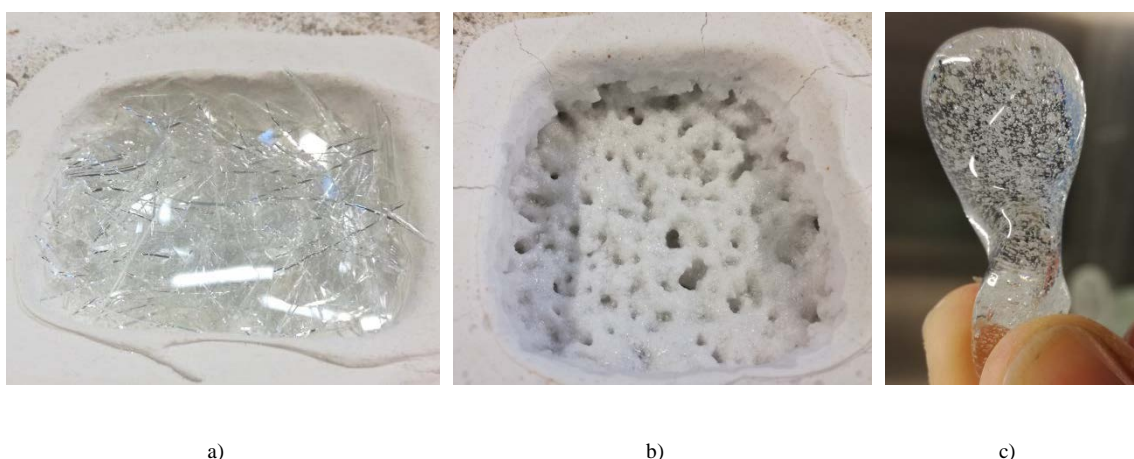


Fig. 15 a), b) Alkali-aluminosilicate glass retrieved from a mobile phone screen before and after firing at 1.250°C, c) Same type of glass melted at 1.500°C and poured into a steel mould.

3.3. Glass combinations

Currently at the recycling plants, a highly automated separation is conducted, strictly excluding any trace of borosilicate, crystal or glass ceramic from the selected soda-lime cullet. Accidental inclusion of these foreign pieces in the batch can cause total glass loss on a float line (NSG 2011) due to the significant differences in their thermal

Cast Glass Components out of Recycled Glass: Potential and Limitations of Upgrading Waste to Load-bearing Structures

expansion coefficient. Ideally though, two or more different recipes of commercial glass should be able to homogenize if kept at a high temperature long enough for new chemical bonds to develop, and then slowly annealed. This research step is a starting point of exploring the possibility of homogenizing different types of glass waste together. This is done gradually, starting by casting similar glass objects and moving towards the combination of different recipes.

- Similar recipes (Clear soda-lime bottles):

The glass recycling industry separates soda-lime containers from numerous different producers into similar colours. The segregated cullet is then successfully recycled together with new raw material. In that sense, casting two different clear bottles together was expected to work well at temperatures corresponding to viscosities of $10^{1.5-2.5}$ poise, which are achieved in the melting tanks. For typical soda-lime silica glasses (Kimble R-6 used as reference), a $10^{2.5}$ viscosity would correspond to 1.254°C (Martlew 2005). Therefore it was opted to test the combination at a lower temperature, namely 970°C . The sample, placed in a flowerpot, was kept at top temperature for 2 hours and then mechanically quenched. A small part of the glass mixture flew down the mould and despite the fact that the glass was fully crystallized, it was homogenized and did not crack during annealing. Regarding the glass mass that remained in the flowerpot, this could be observed by cutting the pot in half. There, zones of crystallized glass of almost same thickness (similar to the thickness of the bottles) are fused together. A few glassy regions are also observed. It is interesting to see that the zone of glass in contact with the flowerpot- which heats up and cools down faster- is more homogeneous and has a different micro-pattern of crystallization. Regardless the above, none of the glass masses cracked, proving that this combination is also feasible at temperatures lower than 1.250°C .



Fig. 16 Results from the kiln-casting of two different types of clear bottles at 970°C . a) Droplets that flowed from the flowerpot down to the mould, b) glass part that fused inside the flowerpot. In this picture, the surface in contact with the flowerpot is seen, c) flowerpot cut in half, showing the layering of the fused glass and the glassy areas within the crystallized mass.

- Similar recipes of different colours (Various soda-lime bottles):
Different colours of wine bottles were used in this experiment, in various sizes from powder to small and medium sized shards. The XRF analysis of 4 different samples showed strong compositional similarity despite the colour shifts. The mix of wine bottles was fired at various temperatures between 940°C -1.450°C. At 970°C the sample would partially fuse and still keep the integrity and colours of the individual pieces. This comes in antithesis with the above mixture of clear bottles (similar recipes to the wine bottles) that homogenized at the same temperature. In this case, the differences in colour/exact recipe, seem to require higher temperatures. Nonetheless, the sample did not crack upon annealing. Adding 10% of fluxing agent (in this case CNa_2O_3) in the powdered mix and firing it at 940°C helped the melting of the top surface but did not lead to a homogenized sample. The sample was homogenized at the tests conducted from 1.200°C and above. Higher temperatures are imperative for the homogenization of this combination.

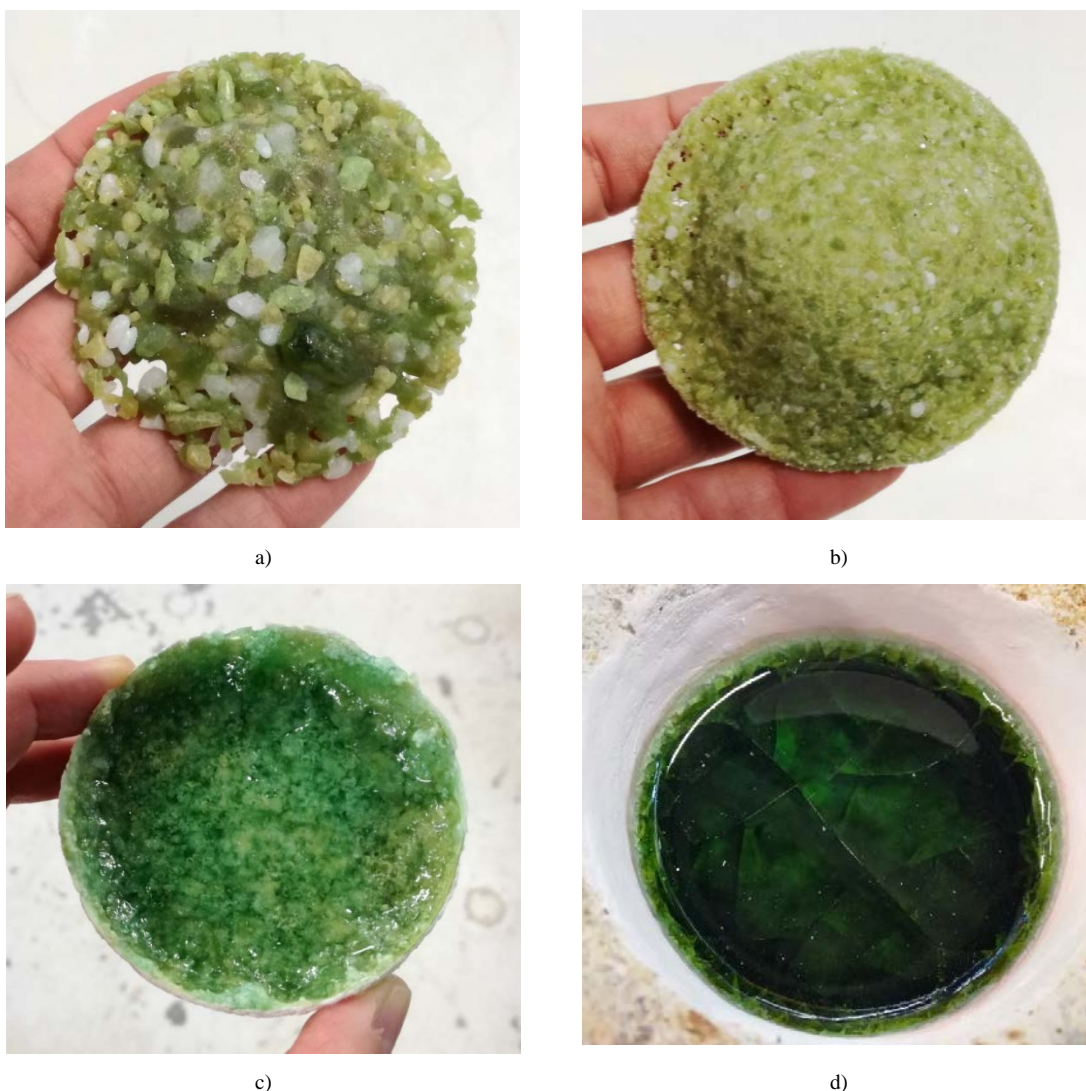


Fig. 17 Samples of different coloured glass bottles kiln-cast at a) 970°C (small shards), b) 970°C (powder), c) 940°C (powder + flux), d) 1.250°C (powder).

- Diverse recipes (Lead crystal and soda-lime glass):
A 1:2 lead crystal to soda-lime shard mixture was prepared and fired at 1.250°C for 3 hours. The crystal shards were placed at the bottom of the silica-plaster mould and the float shards above them. The glass was not stirred during casting. The sample was abruptly cooled down to 625°C and then conservatively annealed from 560°C to 340°C, covering the annealing range of both soda-lime and lead crystal. The resulting glass was extremely clear and without signs of crystallization but it cracked. It is worth to mention that the Crystalcast mould faced a change in structure at such high temperature, becoming from brittle to very rigid. The cracks seen in the glass correspond to the cracks found in the mould, suggesting a force induced by a difference in the thermal contraction of the mould and the glass mixture, rather than a difference within the two different glasses. The glass has also forcefully attacked the wall of the silica plaster mould. This test should be repeated with a different type of mould in order to be able to investigate the feasibility of this combination.

Cast Glass Components out of Recycled Glass: Potential and Limitations of Upgrading Waste to Load-bearing Structures



Fig. 18 Result of the lead crystal and float glass combination, fired at 1.200°C. The cracks of the mould correspond to those found in the glass.

- **Diverse recipes (Borosilicate and soda-lime glass):**
With similar settings as the experiment above, a 1:2 borosilicate to soda-lime shard mixture was prepared and fired at 1.250°C. The borosilicate shards were placed at the bottom of the silica-plaster mould and the float shards above them. An extra step of annealing at 570°C was added to correspond to the borosilicate in the mixture. The resulting glass was clear but more heavily shattered and with partial crystallization close to the wall of the mould. A higher amount of air bubbles were included in the glass. Again the cracks in the mould correspond to the cracks in the glass. A higher temperature and a different type of mould are required for this option to work.

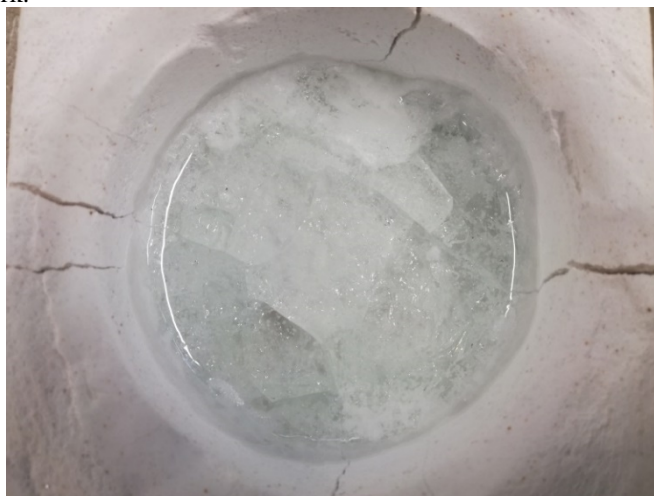


Fig. 19 Result of the borosilicate and float glass combination, fired at 1.200°C. In this case as well, the cracks in the mould correspond with those found in the glass. Areas of crystallization are also observed at the periphery of the glass sample.

4. Conclusions and further research

The results of this research highlight the vast potential of recycling different types of discarded glass into cast glass building components. Products that are almost entirely excluded from the glass-to-glass recycling loop such as CRT panels or crystal tableware- both containing a considerable percentage of heavy metals- proved to be an excellent source of glass that can be kiln-cast at temperatures between 900°C-1.000°C. Soda-lime float, mouth-blown and container glasses, in comparison, needed higher working temperatures ($\approx 1.200^{\circ}\text{C}$) and faster cooling rates to homogenize into transparent glass samples. Yet, their casting at temperatures below 1.000°C combined with a slow cooling, showed an aesthetically and structurally interesting alternative route, that of glass ceramics. The recycling of borosilicate labware can result into very clear glass, and is worth considering despite of the higher than soda-lime required working temperatures. Aluminosilicate glass is considered unsustainable for recycling through casting due to the very high temperature required for it to flow. The limited amount of experiments regarding the mixing of different glass types cannot lead to conclusive data; yet these experiments indicate the potential of combining different glass recipes in castings of higher temperatures. Also, glasses from the same family and of similar colours are considered compatible for cast-recycling.



Fig. 20 The fundamental differences between various types of glass regarding viscosity, thermal expansion and susceptibility to crystallization, introduce challenges to their mixing. Further research should be conducted to overcome the posed obstacles.

In this direction, further investigation will be made on the mixing of different glass recipes with the aim of simplifying the initial, meticulous stage of segregation of the cullet in fewer glass categories. Towards this step, a method should be developed for controlling the quality of the segregated cullet and thus the quality of the final product in terms of strength and aesthetic homogeneity. Future work will be conducted on the mechanical testing of the resulting recycled glass components to define their strength and validate their suitability as building components. Research should also address the recycling of glass contaminated by coatings, laminates, and adhesives, which nowadays constitute a considerable amount of our glass waste. The above steps are necessary for diverting the path of waste glass from the landfills to safe and beautiful structures.



Fig. 21 Successful casting of pieces of (from left to right) float glass, bottle, laboratory tube, computer screen panel, lens, and mouth blown glass, demonstrating the numerous possibilities that arise in creating unique building components.

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Cast Glass Components out of Recycled Glass: Potential and Limitations of Upgrading Waste to Load-bearing Structures

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