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Recycled Glass Mixtures as Cast Glass Components for Structural Applications, Towards Sustainability

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The problem of sustainability represents one of the most important issues that the world has to face nowadays, not only in terms of energy consumption and of the consequent CO_2 emissions, but also in terms of material waste streams that end in landfill. 38 million tons of glass waste are produced every year in the European Union and new targets have been set for 2020 towards a more sustainable management of such wastes. Nowadays, only the container glass industry has reached a considerable recycling rate, while for all the other sectors we are still witnessing downgrading processes. Looking at the world of construction, glass has been more and more employed as a structural material thanks to its high transparency and compression strength. Although the use of glass can be attractive under multiple aspects and its production is continuously increasing, once employed as a construction element, it is rarely reused or recycled due to the high-quality requirement demanded to the industry of production. Nevertheless, besides its main applications as a 2-dimensional element, the new technology of cast glass has been recognised as a potential mean of glass recycling. Here, glass is designed and used under the form of repetitive 3-dimensional units assembled in a whole geometrical shape. In fact, thanks to its higher load-bearing capacity under monolithic shapes, this glass can admit less restrictions and potentially incorporate different types of waste. For this reason, the aim of this experimental work is to find a possible combination between glass families, specifically soda-lime, borosilicate and lead-crystal glass, to be recycled as cast glass components. Each type of glass was powdered or grinded under the form of cullet and different mixtures were prepared to be melted at temperatures of 970°C, 1120°C and 1200°C through the kiln-cast tecnique. Finally, an experimental splitting test was performed to define a force trend and a fracture behaviour for each sample. Some preliminary results have been achieved drawing the guidelines for a further investigation. Soda-lime-silica glass and lead-crystal glass mixture revealed to be the most compliant glass recipe with the required physical and mechanical properties, when reheated at 1120°C. The decrease in the melting temperature of the compound and the higher transparency given by the addition of lead glass revealed the potential benefit, in terms of sustainability, for future projects.

Keywords: Recycling, Cast glass, Sustainability

1. Introduction

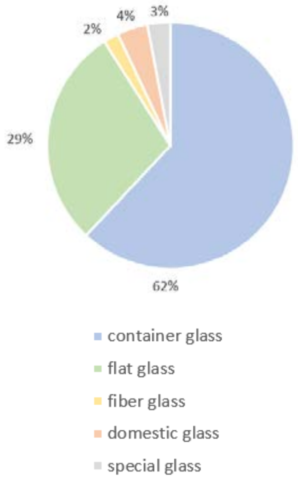
During the last 150 years of industrial evolution it was possible to see only a linear model of production and consumption, where products could just be manufactured, sold, used, and finally discarded as wastes (Ellen Macarthur Foundation 2013). Instead a circular economy enhances the possibility to use fewer resources, lowers environmental impacts and relies less on volatile markets for natural resources (Arup 2011).

Looking at the glass industry and its waste stream, Europe, which can be accounted as one third of the total glass maker (Glass Alliance Europe 2019), can play a key role, and for decades has been engaged to reach higher levels of energy and material efficiency minimizing the environmental impact (Glass Alliance Europe 2019).

Only in 2017, the EU-28 glass production reached a volume of nearly 38 million tons with a global increase of 2,3% compared to 2016 (Glass Alliance Europe 2019). Subdividing the total glass production by different sectors, the container glass industry counts alone for the 62% (figure 1) with about 160 manufacturing plants all over Europe (FEVE 2019). The estimated amount of container glass, only produced in 2017, is 21,452 million tons, but the recycling rate has been reached valuable targets in this sector. The 74% of all glass bottles are indeed collected and recycled, and, each time a ton of glass is recycled, about 580 kg of CO_2 and more than a ton of raw materials are saved along the entire supply chain (Glass Alliance Europe 2018). The second sector is represented by the flat glass industry which counts for the 29% of the total production. Flat glass is manufactured by 7 companies all over the Europe, which sites of production represent the biggest sites of manufacturing with production capacities of up to 850 tons of melted glass per day (Glass Alliance Europe 2017). Although the largest amount of glass derives from the container and flat glass industries, others are the wastes generated (table 1) and, in most of the cases, disposed into landfills, i.e. fiber glass used as reinforcement inside composite materials (around 80% of the total reinforcement fibers available in the market), household's utensils and cookware (domestic glass) and electronic devices screens for TV or PC, which can be counted as special glass.

Glass manufacturing sector	Application or use	Type of glass
Container glass or packaging glass	▪ 75% beverage packaging ▪ 20% other food packaging ▪ 5% packaging cosmetics, pharmaceuticals and technical products	Soda-lime glass
Flat glass	▪ 95% float glass: ○ 75-85% buildings industry ○ 15-25% automotive industry ▪ 5% rolled glass (wired or patterned)	Soda-lime glass
Continuous filament glass fibre	▪ Roving, mat, chopped strand, textile, tissue, milled fibre (90% used for composite materials)	E glass
Domestic glass	▪ Tableware, cookware, vases, ornaments	Soda-lime glass, lead glass
Special glass (CRT not produced in EU anymore)	▪ 75% monitor glass ▪ 25% light bulbs/tubes, ceramic glass, high-temperature domestic glass	Soda-lime, borosilicate glasses

a)



b)

Fig. 1 a) Classification of different industrial sectors (BREF 2009); b) Subdivision of glass production by industrial sector [%] (Glass Alliance Europe 2017).

Table 1: Indices of glass waste production in EU from different industrial sectors.

Type of product	Production [tons/year]	Reference	Amount of waste [tons/year]	Reference
Containers	21.452.000 EU-28	(Glass Alliance Europe 2018)	21.452.000 EU-28	(Glass Alliance Europe 2018)
Flat glass	10.065.000 EU-28	(Glass Alliance Europe 2018)	1.540.704 EU-28	(Hestin, De Veron, Burgos 2016)
Fibre glass	700.000 EU-28	(Glass Alliance Europe 2018)	~ 400.000 EU-27*	(BREF 2009)
Domestic glass	1.253.000 EU-28	(Glass Alliance Europe 2018)	~ 800.000 EU-27*	(BREF 2009)
Special glass (including CRTs)	821.000 EU-28	(Glass Alliance Europe 2018)	~ 500.000 EU-27*	(BREF 2009)

*for these categories of glass waste, data were not available for EU-28.

Of all these types of glass, only the container glass has reached a considerable recycling rate thanks to a well-established close-loop recycling process. For the other glass categories, the highest quality requirement and the impossibility to add other types of waste to the production line have brought to a downgrading process, e.g. recycling glass as aggregate or filler in concrete, as foam or as ceramic-based products (Thomas Dyer 2014).

Considering this scenario, the new technology of cast glass, where glass has been explored and used under the form of repetitive units assembled in a whole geometrical shape (Faidra Oikonomopoulou et al. 2018), has been recognized as a potential mean of glass recycling. Thanks to its higher load-bearing capacity under monolithic shapes, glass can admit less restrictions and incorporate different types of waste. Therefore, different recycling mixtures have been cast at the Glass Lab of the TU Delft, to study the possible combination of glass types from different categories of waste. To validate the possibility of employing such mixtures as cast elements for structural applications, a mechanical splitting test has been performed and reported.

2. Materials

2.1. Types of glass

After evaluating their waste rate and their current recycling processes in addition to their availability, four different types of glass waste were collected (figure 2):

- Soda-lime window glass;
- Borosilicate tubes as shear studs for a pedestrian bridge;
- Lead-crystal glass from domestic utensils;
- Barium glass from Cathode Ray Tubes front panel.

The selected window glass is clear glass without any applied coating. Some studies already showed the low impact of coatings on the recyclability of the material, with exception made for Nickel (Yu 2019). Nevertheless, this selection was made to first study the miscibility of this glass with other glass wastes, without the influence of any applied coating. This opens the possibility to observe during a future research the single effect of the coating on the final mixture.

Table 2 reports the main chemical composition for each type similar to the glass used.

Table 2: Different types of glass chemical composition in wt% (Glass for Europe 2015; James E. Shelby 2005).

Flat glass clear		Borosilicate glass		Lead-crystal glass		CRT front panel	
-		Duran		-		-	
SiO_2	71,8	SiO_2	80,6	SiO_2	55,0	SiO_2	62
Na_2O	13,6	B_2O_3	12,6	PbO	33,0	Na_2O	7,0
CaO	8,78	Na_2O	4,2	K_2O	11,3	PbO	0-2
MgO	3,76	CaO	0,1			K_2O	9,0
Al_2O_3	1,01	MgO	0,05			CaO	<2,0
K_2O	0,60	Al_2O_3	2,2			BaO	2-9
SO_3	0,26	Fe_2O_3	0,04			MgO	<1,0
Fe_2O_3	0,10	Cl	0,1			SrO	10
						Al_2O_3	2,0
						As_2O_3+Sb_2O_3	0,5
						TiO_2+ZrO_2	2,0
						CeO_2	0,2

For soda-lime and lead-crystal glass it was possible to perform an X-ray analysis, and Table 3 shows their main chemical components. This analysis was not conducted for borosilicate glass due to its result inaccuracy on this type of glass.

Table 3: Main chemical composition in wt% of used soda-lime and lead-crystal glass (XRF analysis).

Float glass from IFS-SGT		Leerdam glass	
SiO_2	75	SiO_2	58
Na_2O	12	PbO	29
CaO	08	K_2O	09

Due to the high difference in the chemical composition and physical properties, it was decided to combine only two types of glass (50% each) for each sample with the following combinations:

- Soda-lime and lead-crystal glass
- Soda-lime and borosilicate glass
- Borosilicate and lead-crystal glass
- Soda-lime and barium glass

After being collected, the different types of waste were polished in order to remove surface contaminations and grinded according to the desired dimension. The grinding process was made through a proper machine for 8-10 sec in case of glass powdering and 1-2 sec in case glass was reduced to small pieces of cullet (figure 3). In the second case, a sieving process was required to collect only the pieces between 1 and 3 mm of dimension (figure 4). This specific cullet size has been chosen to reduce the formation of bubbles inside the compound which occurs with powder, and at the same time to avoid the significant thermal expansion which would occurs with larger sizes of cullet pieces.

Finally, the glass wastes were properly mixed and put inside Crystalcast M248 molds to produce cubic specimens of 50 mm in dimensions (figure 5). The kiln-cast technique allowed us to pour directly the glass inside the molds ready for the firing. Different firing schedules were chosen according to the type of mixture: 970°C for 10/20h, 1120°C and 1200°C for 10h. Some samples were produced adding a 10wt.% of Na2O under the form of Na2CO3, in order to reduce the melting temperature and improve the homogeneity of the glass mixture.

a) b) c) d)

Fig. 2 a) Soda-lime glass; b) Borosilicate glass; c) Lead-crystal glass; d) Barium glass.

a) b)

Fig. 3 a) Glass powder; b) Glass cullet.

a) b)

Fig. 4 a) On the left: first sieve used of 1 mm. On the right: second sieve used of 3 mm; b) Glass sieving operation.

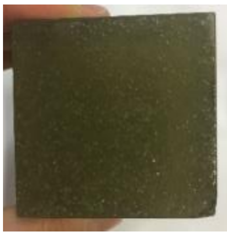
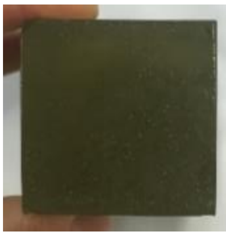
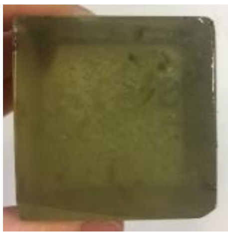
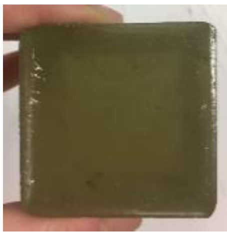
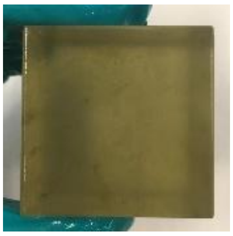
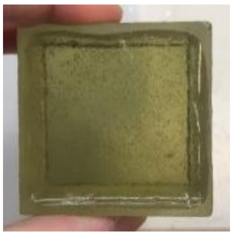
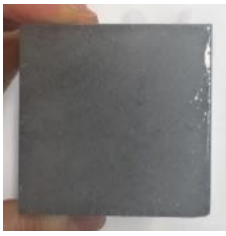
Fig. 5 Molds inside the oven ready for the firing.

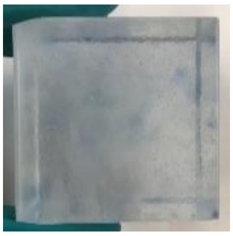
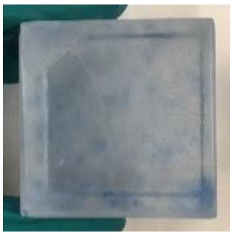
2.2. Glass samples

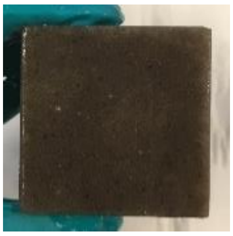
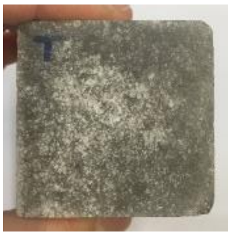
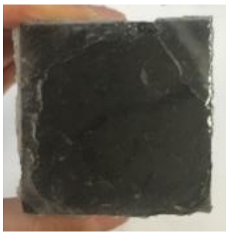
An overview of all the samples produced is reported in table 4. The parameters chosen to characterize the samples are:

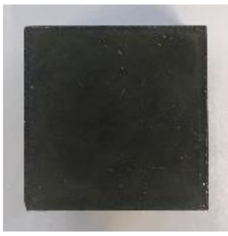
- Mold reaction which indicates the level of interaction between the glass powder/cullet and the surfaces of the mold;
- Cracks presence caused by the different thermal expansion coefficient between the glass types;
- Bubbles level;
- Bubbles dimension;
- Contamination caused by the powdering process of other materials (metal and stones) inside the grinding machine.

Table 4: Samples overview.

SAMPLE CODE	SL-B01	SL-C01	SL-E01
Mixture		Soda-lime & lead-crystal	
Fluxing agent	no	yes	yes
Firing T.	970°C	970°C	970°C
			
Mould reaction	Absent	Medium	Medium
Cracks presence	Absent	Absent	Absent
Bubbles level	Hight	Medium-high	Medium
Bubbles dimension	Medium	Small	Small
Contamination	Medium-high	Medium-high	Medium-high
SAMPLE CODE	SL-A02	SL-H01	SL-I02
Mixture		Soda-lime & lead-crystal	
Fluxing agent	no	no	no
Firing T.	1120°C	1120°C	1120°C
			
Mould reaction	Absent	Absent	Absent
Cracks presence	Absent	Absent	Absent
Bubbles level	Low	Low	Low
Bubbles dimension	Small	Small	Small
Contamination	Medium-high	Medium	Medium
SAMPLE CODE	SL-H02	SL-F01	L-E02
Mixture	Soda-lime & lead-crystal		Lead-crystal
Fluxing agent	yes	no	no
Firing T.	1120°C	1120°C	970°C
			
Mould reaction	High	Absent	Absent
Cracks presence	Absent	Absent	Absent
Bubbles level	Medium	Low	High
Bubbles dimension	Small	Small	Small
Contamination	Medium	Low	Medium-high

SAMPLE CODE	SL-ST01	SL-ST02	SL-ST03
Mixture		Soda-lime & lead-crystal	
Fluxing agent	no	no	no
Firing T.	1120°C	1120°C	1120°C
			
Mould reaction	Absent	Absent	Absent
Cracks presence	Absent	Absent	Absent
Bubbles level	Low	Low	Low
Bubbles dimension	Small	Small	Small
Contamination	Low	Low	Low
SAMPLE CODE	S-D01	SB-A01	SB-G01
Mixture	Soda-lime	Soda-lime & borosilicate	
Fluxing agent	no	no	no
Firing T.	970°C	1120°C	1200°C
			
Mould reaction	Low	Low	Medium
Cracks presence	Absent	Absent	Absent
Bubbles level	High	High	High
Bubbles dimension	Small	Big	Big
Contamination	Medium-high	Medium-high	Medium-high
SAMPLE CODE	SB-F02	SB-G02	BL-I03
Mixture	Soda-lime & borosilicate		Borosilicate & lead-crystal
Fluxing agent	no	no	no
Firing T.	1120°C	1200°C	1120°C
			
Mould reaction	Medium	Medium-high	Medium-high
Cracks presence	Absent	Low	Not visible
Bubbles level	Not visible	Not visible	Not visible
Bubbles dimension	-	-	Not visible
Contamination	Absent	Absent	Absent
SAMPLE CODE	B-I01	BL-A03	BL-F03
Mixture	Borosilicate	Borosilicate & lead-crystal	
Fluxing agent	no	no	yes
Firing T.	1120°C	1120°C	1120°C

SAMPLE CODE	B-I01	BL-A03	BL-F03
			
Mould reaction	Medium	Medium-high	Medium-high
Cracks presence	Absent	Not visible	Medium
Bubbles level	High	High	High
Bubbles dimension	Small	Small	Small
Contamination	Medium-high	Medium-high	Medium-high

SAMPLE CODE	SBA-H03
Mixture	Soda-lime & barium glass
Fluxing agent	no
Firing T.	1120°C
	
Mould reaction	Medium
Cracks presence	Absent
Bubbles level	High
Bubbles dimension	Medium-small
Contamination	Medium-high

As a first consideration, the obtained results are particularly affected by the conditions and limitations of molds and material production as well as strictly related to the established firing settings. Powder contaminations and the interaction with the mold are assumed to have potentially changed the properties of the final materials. For this reason, it is suggested the use of a grinding machine not employed for other recycled materials, such as stones or metals, and of a proper steel mold to avoid any possible interaction with the glass powder.

Given these considerations, the following observations can be made:

Soda-lime window glass, when melted again under the form of powder at the relatively low temperature of 970°C, changes its amorphous phase becoming a crystallized porous material.

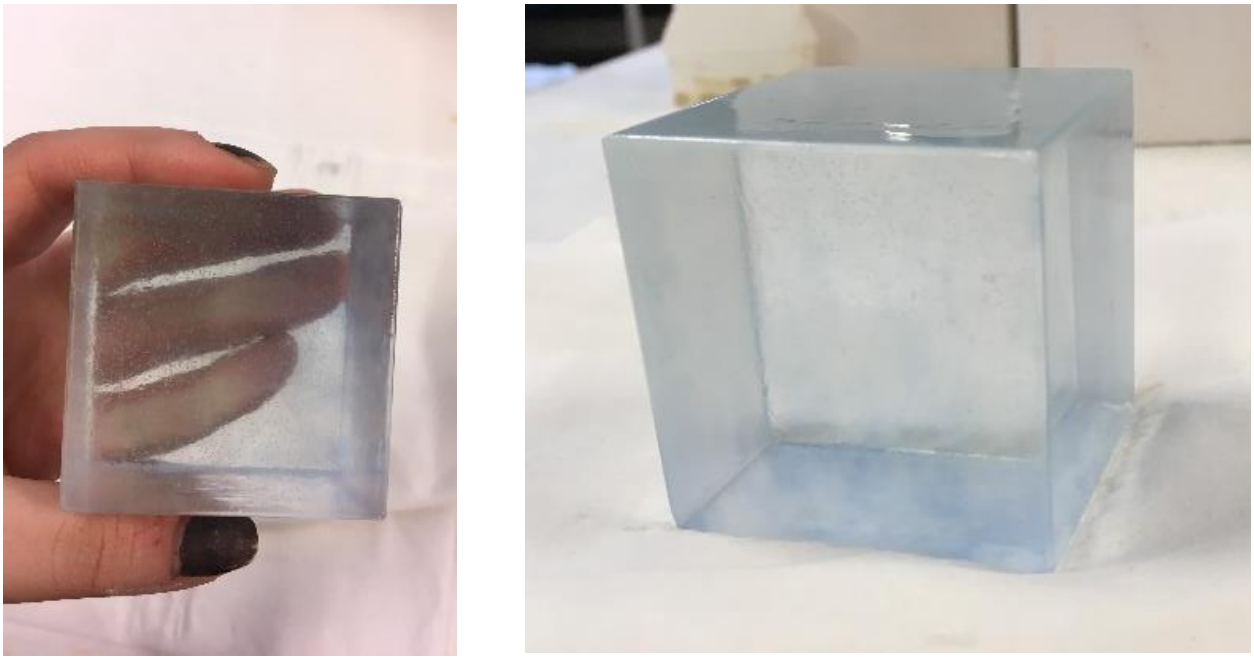
Although its powder mixture with borosilicate glass results in a crystallized material, when fired at 1200°C, it shows a lower density and a quite homogenous bone structure, which makes this compound potentially suitable for thermal insulation applications. On the contrary, the same mixture under the form of cullet generates a non-homogenized material, where the two glass types melt as separate phases.

Lead-crystal glass lowers significantly the melting temperature of soda-lime window glass, both under the form of powder and cullet, and gives a glass body with the required transparency when no contamination occurs (figure 6). The lower melting temperature can be explained by the higher ionic radius of Pb^{2+} inside lead glass, which makes less strong the network links inside the chemical structure (James E. Shelby 2005). Although the glass mixture with the most promising result is the one fired at 1120°C, it is proved that the same glass mixture can be fired at a lower temperature (970°C) using a 10 wt.% of fluxing agent (Na_2CO_3).

Lead-crystal glass acts as a fluxing agent also when mixed with borosilicate glass, but the higher difference in the thermal expansion coefficient brings to cracking phenomena, when the sample is fired at 1120°C under the form of powder. Moreover, the different density is resulting in the phase separation of the two glasses, especially when melted under the form of small cullet.

Given the most promising mixture between soda-lime window glass and lead-crystal glass, an attempt was made substituting the lead glass with barium glass deriving from TV front panel (CRT glass). The experiment turned out in a glass body, but the main limitation is represented by the dark color of the CRT product and by the higher amount of bubbles inside the glass matrix.

Finally, as almost recognized in all the samples, the increase of the initial size of the glass waste reduces the contamination level and the amount of bubbles entrapped inside, but also contrasts the homogenization of the final material.



a) b)

Fig. 6 a), b) Soda-lime and lead-crystal glass mixture after being polished.

3. Splitting test

3.1. Set-up

Table 5. shows in order all the specimens tested and reports the surface treatment to which each sample was subjected before the test.

Table 5: Specimen classification.

Specimen number	Sample code	Description	Surface treatment
1	SL-A02	Soda-lime - lead-crystal (powder) 1120°C	Polishing at 200-grit
2	SL-B01	Soda-lime - lead-crystal (powder) 970°C	Polishing at 60-grit
3	SL-C01	Soda-lime - lead-crystal - fluxing agent (powder) 970°C	Polishing at 200-grit
4	SL-E01	Soda-lime - lead-crystal - fluxing agent (powder) 970°C	Polishing at 60-grit
5	SL-F01	Soda-lime - lead-crystal (cullet) 1120°C	Polishing at 200-grit
6	BL-A03	Borosilicate - lead-crystal (powder) 1120°C	Polishing at 60-grit
7	SB-F02	Borosilicate - soda-lime (cullet) 1120°C	Polishing at 60-grit
8	SB-G02	Borosilicate - soda-lime (cullet) 1200°C	Polishing at 60-grit
9	L-E02	Lead-crystal (powder) 970°C	Polishing at 60-grit
10	SBA-H03	Soda-lime - barium (powder) 1120°C	Polishing at 60-grit
11	S-D01	Soda-lime (powder) 970°C	Polishing at 60-grit
12	SB-G01	Borosilicate - soda-lime (powder) 1200°C	Polishing at 60-grit
13	SB-A01	Borosilicate - soda-lime (powder) 1120°C	Polishing at 60-grit
14	SL-ST01	Soda-lime - lead-crystal (cullet) 1120°C	Polishing at 600-grit

Specimen number	Sample code	Description	Surface treatment
15	SL-ST02	Soda-lime - lead-crystal (cullet) 1120°C	Polishing at 600-grit
16	SL-ST03	Soda-lime - lead-crystal (cullet) 1120°C	Polishing at 600-grit
17	SL-I02	Soda-lime - lead-crystal (powder) 1120°C	Polishing at 200-grit
18	B-I01	Borosilicate (powder) 1120°C	Polishing at 60-grit
19	BL-I03	Borosilicate - lead-crystal - fluxing agent (cullet) 1120°C	Polishing at 60-grit

The splitting test set-up is shown in figure 7.

The test was performed through a Zwick Z100 displacement-controlled machine with a displacement rate of 0,2 mm/min. The specimen was attached to the steel plate through a duct tape, first on the upper surface in the axial force direction and, later, on the top and bottom surfaces to give a higher stability.

An initial error due the presence of the tape on the contact surface was admitted.

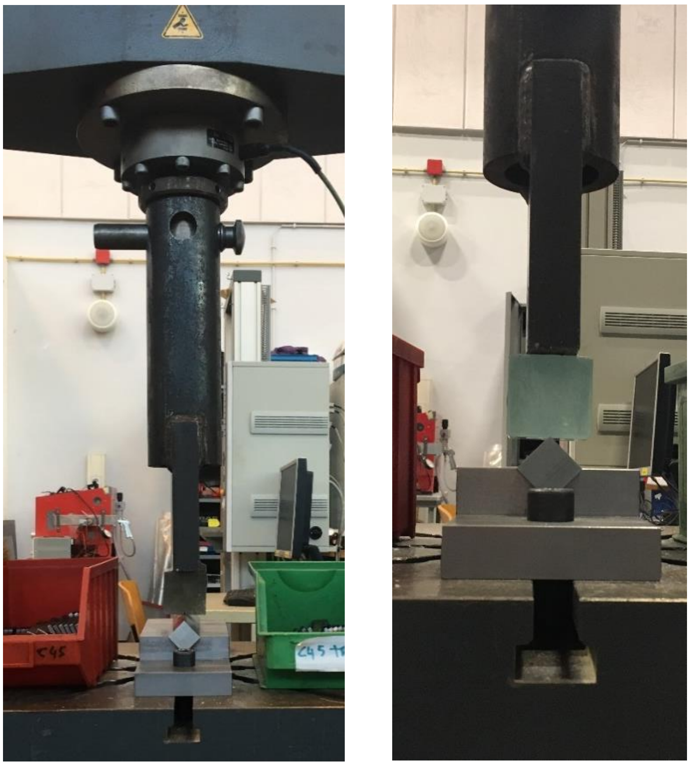


Figure 7. Splitting test set-up.

3.2 *Results*

Table 6 shows an overview of the results in terms of Fmax and corresponding displacement.

Table 6: Mechanical test results.

Test number	F_{max} [N]	dL at F_{max} [mm]
1	18393,87	1,10
2	18417,48	2,32
3	9776,24	0,64
4	22360,04	0,77
5	22034,46	0,81
6	15412,53	1,02
7	4478,62	0,79
8	6460,17	1,53
9	13701,48	0,74

Test number	F_{max} [N]	dL at F_{max} [mm]
10	20607,31	0,92
11	11149,03	1,26
12	13445,31	1,40
13	4710,61	1,11
14	25857,15	1,23
15	27568,41	1,24
16	32258,35	1,37
17	23965,23	1,15
18	25619,02	1,22
19	5761,71	0,93

The force-displacement diagrams are reported and subdivided according to the force trend (figure 8).

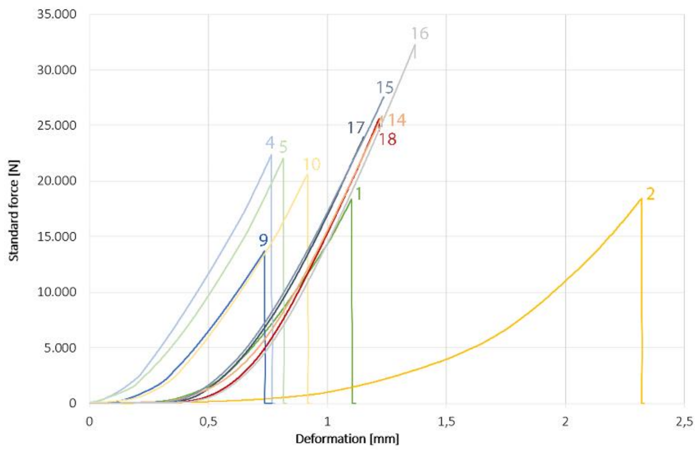
Three main trends can be identified:

a) a continuous trend, where the force increases continuously until reaching the fracture point;
b) an indented trend before the maximum peak, where the force increases discontinuously until reaching the fracture point;
c) an indented trend after the maximum peak, where the force decreases discontinuously after the fracture point.

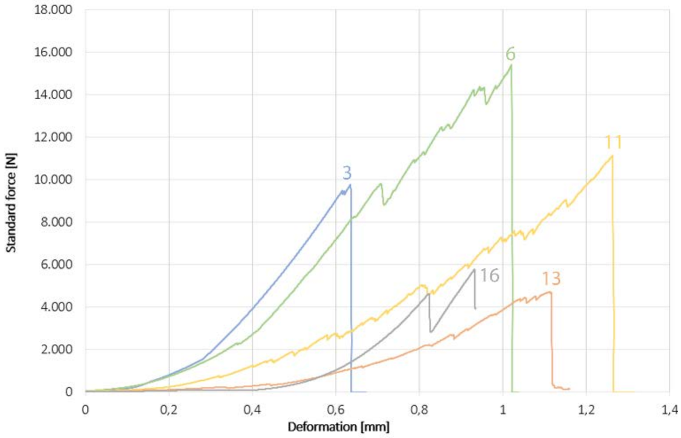
The first trend is mainly recognized for those samples presenting a homogeneous glass structure, whilst the second trend is shown by partially foamed samples with a higher porosity, and the third one by partially crystallized samples where the two types of glass did not properly homogenize (figure 9).

The lowest resistance, with a maximum force in the range of ~ 4 - 13 kN, mainly appears for the second and third trend indicating the porosity and the non-homogeneity of the material as strong affecting parameters. Especially for partially crystallized specimens, the low range of applied load is related to the unmixed glass particles inside the material with a resulting concentration of stresses on the small interfaces.

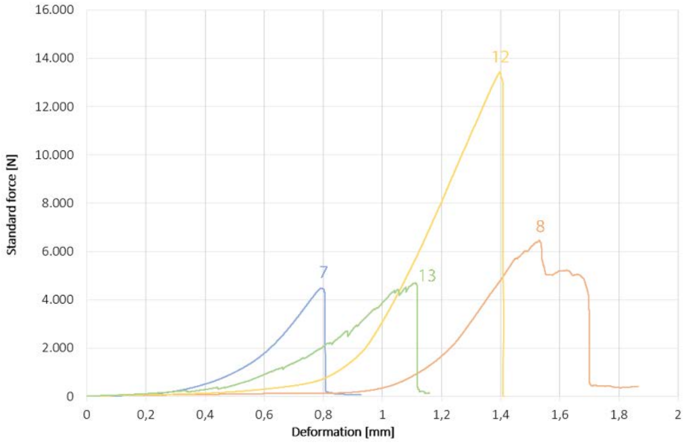
On the contrary, the glass specimens show good values of resistance reaching force peaks between 20 and 35 kN. The fracture behavior is well visible from the broken surfaces of the specimens, where the waves propagation starts from the angles of the sample suggesting a concentration of stresses in these areas (figure 10).



a)

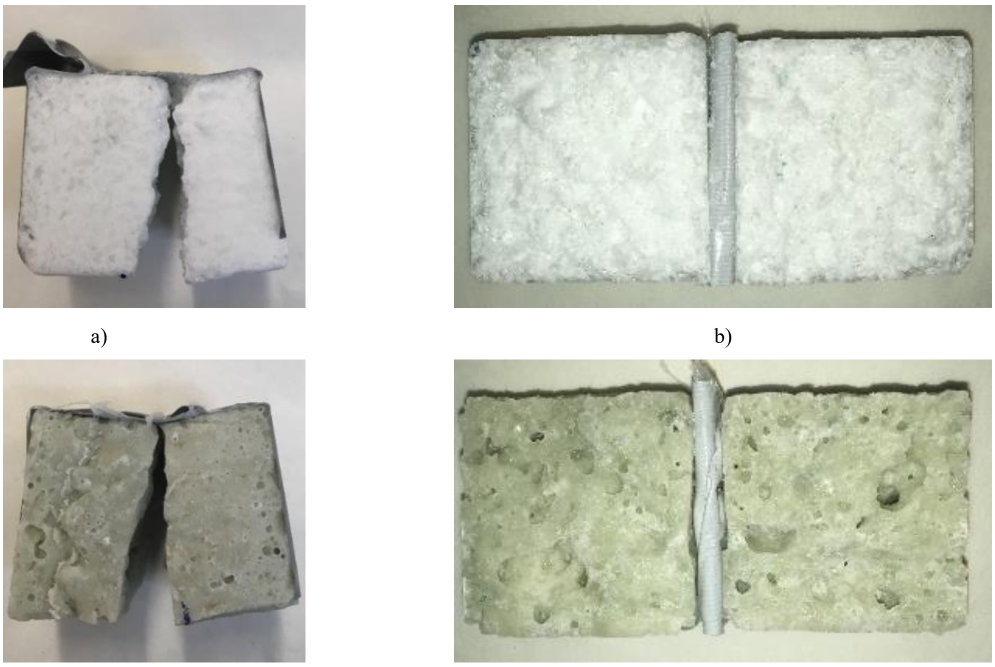


b)



c)

Figure 8. Force-displacement diagram of specimens with: a) the first force trend; b) the second force trend; c) the third force trend.



a) b)

c) d)

Fig. 9 a) b) Fracture surface of specimen 12; c) d) Fracture surface of specimen 7.

a) b)

Fig. 10 a) b) Fracture surface of specimen 16.

4. Conclusions

The aim of this experimental work is to find a possible combination between glass compounds to be recycled as cast glass components for structural applications. The initial input was to rethink glass as a construction material in a more sustainable approach. Different attempts have been performed showing the potentiality of some mixture and drawing the guidelines for a future research.

The experiments performed in the laboratory showed a clear correlation between the results achieved and the working facilities and conditions. More precisely, glass powdering was affected by stone and metals contamination present inside the grinding machine, which was supposed to have changed the final properties of the glass mixture. Moreover, some compounds were reacting with Crystal Cast molds, especially in presence of the fluxing agent NaCO3, with a consequent adhesion between the mold and the specimen, and the change of its surface properties.

At the given conditions, the following main achievements have been reached:

- Soda-lime-silica glass and lead-crystal glass mixture revealed to be the most compliant glass recipe with the required physical and mechanical properties for a cast glass component in structural applications. The result was particularly promising due to the lower melting temperature of 1120°C with respect to the one of soda-lime window glass. Moreover, the same glass mixture was proved to be fully homogenized at the lower temperature of 970°C with an additional 10 wt.% of fluxing agent. Another important aspect under consideration was the transparency given by lead-crystal glass to the mixture. Soda-lime window glass starts indeed to show greenish coloring at thickness higher than the ones employed in the float process, due to its ferric oxide content (in the range of 0,1%). Adding lead glass, its final transparency is improved.
- Other mixtures give homogenized material with a different reorganized structure. Soda-lime window glass mixed with borosilicate glass turned out in a porous crystallized material with an acceptable resistance value, when melted at the higher temperature of 1200°C under the form of powder. Although its chemical structure does not give a transparent material, its high porosity lowers its density giving to the material a higher insulation property. For this reason, such compound could be further studied as a potential insulation material.

Further research will be focused on:

- Both XRF and SEM analysis on the obtained mixture to observe the local chemical composition.
- Extrapolating the tensile strength of the material with the new set-up used for the splitting test. A FEM model should be completed.
- The production of prototypes with soda-lime and lead-crystal glass mixture and the following mechanical testing.
- More experiments on other glass mixtures, for example soda-lime with borosilicate glass, to be employed for same or different applications.
- The use of window glass with low-E coatings to evaluate their possible effects on the final mixture with other types of glass waste.

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