Innovative use of waste CRT glass in wall cladding tiles production

DANUTA CHMIELEWSKA[1], ROMAN GEBEL[1], BARBARA SYNOWIEC[1],
ZBIGNIEW JAEGERMANN[1], ANDRZEJ ŁOSIEWICZ[1], AGNIESZKA DUCKA[1]
OTTO ANDERSEN[2]
[1]Institute of Ceramics and Building Materials,
02-676 Warsaw, Postępu 9,
POLAND,
info@icimb.pl http://www.icimb.pl

[2]Western Norway Research Institute Vestlandsforsking
Fosshaugane Campus - Røyrgata 4, 6851 Sogndal
NORWAY
otto.andersen@vestforsk.no http://www.vestforsk.no

Abstract: -This poster presents the results of implementation of the project entitled "Recycling of CRT glass waste in the production process of glass tiles – determination of production process parameters" as a part of Polish-Norwegian Research Programme. The objective of this project was to develop a new production technology of glass tiles made exclusively from recycled CRT glass. The results of natural radioactivity testing of waste CRT glass confirmed that it can be used as a raw material in the production of building materials.
Testing of physical and chemical properties of waste CRT screen and cone glass was carried out to determine the manufacturing process parameters. Tests of physical and chemical properties were performed for materials prepared in a pilot scale according to the standards applicable to ceramic tiles. The resulting parameters determine the application of CRT glass tiles as a cladding material for indoor walls.

Key-Words: - CRT glass, glass tiles, glass recycling, CRT glass processing,

1 Introduction

Every year, more than ten thousand tons of CRT glass finds its way to city landfills, and while kinescope television sets and screens are being withdrawn from use, this is going to continue for the coming years (estimated 5 to 7 years). Accumulation of this type of waste is not without consequence for the environment.
The Directive 2002/96/EC on waste electrical and electronic equipment (WEEE) requires Member States to collect, process, recover and recycle this waste. The objectives of this Directive include carrying out studies to develop innovative measures to prevent environmental degradation.
The Institute of Ceramics and Building Materials (ICiMB) has therefore focused on CRT waste glass utilization to promote actively this strategy. It has carried out the project entitled "Recycling of CRT glass waste in the production process of glass tiles – determination of production process parameters" as a part of Polish-Norwegian Research Programme.
The Promoter of the Project is the Institute of Ceramics and Building Materials, located in Warsaw. Stiftinga Vestlandsforsking/Western Norway Research Institute, located in Sogndal, is a Partner in the Project.

2 Problem Formulation

The objective of the project was to develop a new production technology of glass tiles made exclusively from recycled CRT glass.

2.1 Scope of work

The scope of research included:

1. Determination of physical and chemical properties of kinescope glass in relations to screen glass and cone glass.
2. Development of elementary process parameters for glass tile manufacturing technology.
3. Manufacturing a pilot batch of glass tiles in semi-technical scale.
4. Test of tiles properties according to EN 14411:2009 "Ceramic tiles. Definition, classification, properties and marking".

3 Problem Solution

3.1 Determination of physical and chemical properties of CRT glass in relations to screen glass and cone glass.

Testing of physical and chemical properties was performed on a waste glass obtained from the Polish plant, where CRT glass waste is collected, and a waste glass, which had been delivered by the Norwegian Partner.

Table 1. Physical and chemical properties of the CRT glass waste

Test Type	Polish glass		Norwegian glass	
	Screen	Cone	Screen	Cone
Determination of lead content %	0,196	21,10	0,233	21,77
Characteristic melting points °C :				
Shrinkage starting temperature	574	565	614	580
Horns temperature	771	698	775	690
Barrel temperature	795	740	800	755
Ball temperature	870	801	830	790
Hemisphere temperature	980	890	925	870
Melting temperature	1046	965	1006	940
Expansion Coefficient α $\Delta L/(L_0 \times \Delta T) \times E^{-6}$ 25-410°C	8,3274	8,3514	8,3548	8,3593
Natural radioactivity Bq/kg*:				
Activity indicator f1	1,08	0,07	no data	no data
Activity indicator f2	85,18	15,25	no data	no data

*Maximum allowable values: f1=1,2 Bq/kg, f2=240 Bq/kg .

The test results admit the possibility of applying the tested material as raw material for manufacturing of CRT glass tile, because f1 and f2 values are below the maximum permitted ones.

3.2 Development of elementary process parameters for glass tile manufacturing technology.

The tests were performed according to the following, established initial scheme of CRT glass tile production:

1. Grinding raw materials in a ball mill,
2. Drying the suspension in a spray dryer,
3. Pressing tiles using granulate powder,
4. Firing green tiles in a roller furnace,

Determination of manufacturing process parameters was performed on a waste glass obtained from the Polish plant, where CRT glass waste is collected (due to the availability of large amounts of the material).

3.2.1. Conditions for grinding in a ball mill

Parameters of size distribution affect the pressing process and the associated functional strength of the pressed material as well as the final result after firing (Table 2). In order to determine the effect or to finally determine the optimum size distribution parameter D (v, 0.9) grinding samples were performed for the assumed mass composition (mass composition is the subject of the future patent application), for the resulting pressed tile material in the form of a cross-section of 60x60 mm with a pressure applied of 85 kN, pressing pressure set at 23.44 MPa.

Table 2. Final results of grinding

Test No	D(v,0.1) µm	D(v,0.5) µm	D(v,0.9) µm	Notes
1	0.35	11.40	49.98	Granules well pressed, lack of functional strength of the tile
2	0.33	9.15	41.70	
3	0.30	8.36	40.65	
4	0.34	7.08	35.47	Granules well pressed, poor functional strength of the tile
5	0.46	7.55	31.11	Granules well pressed, good functional strength of the tile
6	0.47	8.05	28.52	
7	0,58	7,27	24,81	

The milling process was performed in an industrial ball mill for wet grinding, while maintaining the following grinding specification: 1 part by weight of waste glass and 1 part by weight of grinding media and 0,6 part by weight of water.

3.2.2 Drying and granulate

The process tests were conducted on the spray Dorst dryer (spray nozzle 1 mm). The tests consisted in choosing the drying temperature ensuring obtaining well pressed granules, while, having good functional strength of the pressed tiles. Spray drying tests were carried out at three temperatures of inlet gases: 350°C, 300°C, 288°C.

The flexural strength was measured in the raw state. Values of 1,95 MPa, 1,23 MPa and 0,27 MPa were achieved. The pressed samples had good functional strength and, after firing at 810°C had sharp edges and showed zero absorption.

A sample was made on a semi-technical scale of preparation of granules with two suspensions of varying density.

During the final test Dorst dryer was operating under temperature ranges: top 315-400°C, down 105-120°C. Suspension density assumed was between 1,52 and 1,6 g/cm^3.

3.2.3 Pressing

Samples of pressing were carried out on a semi-automatic Dorst TPA45 press.

For tiles with a cross section of 60x60 mm pressing force of 85 kN was used, and for tiles with a cross-section of 117,2x 117,2 mm, 322kN pressing force was applied. Pressure of pressing was set at 23 MPa. Humidity of the granulate fed to the press was in the range of 5,4- 6,5%.

3.2.4 Sintering

Tiles of cross-section 60x60 mm were fired in an electric laboratory roller furnace made by Italian company Ceramic Instruments at three temperatures: 790°C, 800°C, 810°C. Profilometric tests were carried out for sintered tiles. The test results are presented in the Table 3.

Table 3. Results of profilometric tests

Sintering temperature	Ra [μm]	Rz [μm]
790°C	6,22	295,1
800°C	6,40	296,9
810°C	3,17	176,6

Firing temperature 810° results in a significant reduction in roughness parameters.

A fired tile was subject to quantitative and qualitative analysis of the phase composition. The tests result indicates the presence of the amorphous phase in the amount of 98,43 ± 0,078% by weight and quartz in the amount of 1,57 ± 0,074% by weight.

The firing process has been tested in operating conditions of industrial electric furnace and a gas furnace.

The tests for maximum firing temperature were carried out in the following temperature ranges: 790-810°C and 900-920°C (temperatures presented in Table 4 refer to tests conducted in the electric furnace).

Table 4. Process parameters

Milling	1. D parameter (v,0.9) in range of 32-24 μm 2. Outflow time from the Ford cup nr 5- about 19 seconds
Granulation	1. Suspension density: 1,52-1,6 g/cm^3 2. Granulate moisture: in range of 6-8%
Pressing	1. Pressing force for given mold cross-section: - 60x60mm – 85 kN - 117,2x117,2mm – 322 kN 2. Pressing pressure: 23 MPa
Sintering	1. 800-810°C 2. 900-910°C

3.3. Manufacturing a batch of glass tiles in semi-technical (pilot) scale.

According to the findings of the earlier tests, grinding of 200 kg glass screen was performed in a ball mill for 20 h to get the particle size distribution shown in the table 5.

Table 5. Particle size distribution of CRT glass used for pilot batch

Conc. = 0.0161 %Vol	Density = 1.000 g/cm^3		S.S.A.= 7.1558 m^2/g
Distribution: Volume	D[4, 3] = 10.96 um		D[3, 2] = 0.84 um
D(v, 0.1) = 0.26 um	D(v, 0.5) = 5.26 um		D(v, 0.9) = 29.92 um
Span = 5.640E+00	Uniformity = 1.797E+00		

Size (um)	Volume Under%	Size (um)	Volume Under%	Size (um)	Volume Under%	Size (um)	Volume Under%
0.05	0.00	0.58	20.23	6.63	54.42	76.32	99.70
0.06	0.11	0.67	21.83	7.72	57.55	88.91	99.96
0.07	0.35	0.78	23.38	9.00	60.86	103.58	100.00
0.08	0.71	0.91	24.99	10.48	64.34	120.67	100.00
0.09	1.20	1.06	26.70	12.21	67.98	140.58	100.00
0.11	1.84	1.24	28.53	14.22	71.77	163.77	100.00
0.13	2.65	1.44	30.48	16.57	75.70	190.80	100.00
0.15	3.65	1.68	32.51	19.31	79.66	222.28	100.00
0.17	4.89	1.95	34.60	22.49	83.51	258.95	100.00
0.20	6.40	2.28	36.74	26.20	87.14	301.68	100.00
0.23	8.22	2.65	38.93	30.53	90.40	351.46	100.00
0.27	10.31	3.09	41.20	35.56	93.20	409.45	100.00
0.31	12.52	3.60	43.56	41.43	95.47	477.01	100.00
0.36	14.68	4.19	46.04	48.27	97.20	555.71	100.00
0.42	16.66	4.88	48.67	56.23	98.43		
0.49	18.51	5.69	51.46	65.51	99.23		

A control samples of pressing and firing were done. A positive result was obtained. The tiles had good functional strength after pressing and retained edges being fired at 805°C. Tested tiles was assessed positively.

The ground suspension was divided into 5 parts to produce colorless, brown, green, blue and black tiles. The suspension was stained with pigments produced by the Department of Decoration Agents of the Institute of Ceramics and Building Materials. The pigments used in an amount of 2% relative to the dry matter in the slurry were listed in table 6.

Table 6. Pigments used in pilot batch

No	Pigment	Symbol	Elemental Composition
1	brown	26004	Zn,Fe,Cr
2	black	14808	Co,Ni,Fe,Cr
3	green	11131	Co, Cr
4	blue	12259	Zn, Co, Al., Cr

Pilot batch of CRT glass tiles were manufactured on a semi-technical scale in the workshop conditions of CERAMIKA PARADYZ industrial plant located in Opoczno, Poland. Tiles were fired at two different temperatures to test the possibility of obtaining semi-gloss tiles with sharp edges (Fig.1) and gloss tiles with rounded edges (Fig.2). In both cases, the firing process took place at a temperature 150 to 250°C lower than the firing temperature of ceramic tiles. The tiles were colored in-mass with the use of ceramic pigments in order to improve the final aesthetical effect.

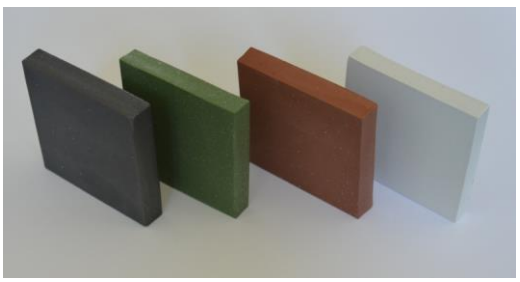

Fig.1. Semi-gloss tiles with sharp edges

Fig.2. Gloss tiles with rounded edges

3.4. Tests of tiles properties according to EN 14411:2009 "Ceramic tiles. Definition, classification, properties and marking"

Since a glass tile produced by sintering of glass powder is a completely new product, there are no applicable standards yet. To evaluate its application properties appropriate tests were performed according to the European standard applied to ceramic tiles. As this product is a dry pressed, unglazed tile, by analogy with ceramic tiles, it was classified to B1 Group of the standard. The testing was conducted in the Research Laboratory of the Institute of Ceramics and Building Materials (Notified body No. 1487).

The parameters of prepared CRT glass cladding tiles were examined based on the following standards:

1. Average permeability - PN-EN ISO 10545-3:1999
2. Flexural strength - PN-EN ISO 10545-4:2012
3. Breaking strength - PN-EN ISO 10545-4:2012
4. Deep abrasion – PN-EN ISO 10545-6
5. Chemical resistance for unglazed tiles - PN-EN ISO 10545-13:1999 + Ap1:2003
6. Frost resistance – PN-EN ISO 10545-1

Table 7. Parameters of cladding tiles manufactured in pilot scale.

Parameter	Value obtained	Value required
Average Permeability [%]	0,4	≤0,5
Flexural strength [N/mm^2]	≥ 46	min. 35
Breaking strength [N]	≥2081	min. 1300
Deep abrasion [mm^3]	≤175	263
Chemical resistance for unglazed tiles	no visible change	declared
Frost resistance	does not meet the standards	

4 Conclusion

1. The resulting parameters determine the application of CRT glass tiles as a cladding material for indoor walls.
2. The technology is energy-efficient compared to the conventional technology of manufacturing cladding tiles.
3. The developed solution makes it possible to fully utilize waste CRT glass.
4. Utilization of waste glass will contribute to the reduction of extraction of fossil fuels.