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WASTE LIQUID CRYSTAL DISPLAYS (LCDS) GLASS AS AGGREGATE SUBSTITUTE IN CONCRETE PRODUCTS

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Abstract

The increasing amount of waste electrical and electronic equipment (WEEE) is one of today's concerning problems. Due to every day's increasing quantities of WEEE, different methods have been developed to achieve the most efficient extraction and usage of valuable components such as metals, plastics, glass, etc. In this paper, concrete C20/25 was prepared with waste liquid crystal display (LCD) glass used as a replacement for fine aggregate in percentages 1%, 5% and 10%. For comparison purposes concrete with replacement level of 0% was prepared. First set of replacements was done with grinded LCDs (d≤ 10 mm), while in other set were used LCD glass residues after metal extraction. Properties of fresh and hardened concrete were observed. This research indicated higher values of compressive and tensile strength of concrete with treated LCDs as opposed to concrete with untreated LCD for all replacement percentages, e.g. for 1% replacement compressive strength was 33.4MPa as opposed to 35.53MPa, and for tensile splitting strength 2.56MPa opposed to 3.75MPa in concrete with 1% replacement with treated LCDs. Results indicate 1% and 5% are appropriate percentages of replacement.

Key words: circular economy, concrete, recycling, waste liquid crystal displays

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1. Introduction

The growing dependence on electric and electronic equipment (EEE) in everyday life has highlighted a new environmental question, i.e. how to manage with the fastest growing waste stream (Charles et al., 2017). Since EEE contains a wide range of compounds, some of which are toxic or hazardous, it also contains base and precious metals. Many European policies point out recovery of scarce resources. The European Union (EU) has adopted an ambitious Circular Economy (CE) package (Ferella et al., 2017). Among EEEs, nowadays are widely used liquid crystal displays (LCDs); in TVs, laptops, desktops and any other device coupled with a screen. Waste LCDs are classified as waste electronic and electric equipment (WEEE) (Ferella et al., 2017).

The CE package aims to "close" the loop of product lifecycles through improved product design and improved waste management. Maximizing the recovery of materials from End-of-Life (EoL) products and designing greener products are vital for CE package as stated in (Van Schaik & Reuter, 2016). Based on the production of mobile devices and the future demand, LCD production is predicted to continuously increase and thus also the amount of waste LCD (Kim et al., 2017).

LCDs have a complex composition. The components of the LCD panel include glass (85–87%), polymer membrane (12.7–14%), and liquid crystals (0.12–0.14%). A liquid crystal is composed of glass substrates, liquid crystal, indium-tin-oxide (ITO) conductive glass and black matrix (chromium oxide) (Góra et al., 2019). ITO electrode as a vital component

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of LCDs consumes roughly 90% of annual production of indium. The indium content in ITO glass powder from waste LCDs could reach 0.0576% which is much higher than that in indium-containing ores (less than 0.01%). As a result, the recovery of indium from electronic waste is the main source of indium production, but recovery cost limits industry applications (Cui et al., 2020). The use of recycled LCD glass in concrete and ceramics production is an example of “closing the loop” in recycling (Wang and Huang, 2010).

Concrete is heavily used as a construction material in modern society and the demand for virgin natural resources is increasing day by-day (Aprianti, 2017; Luhar and Luhar, 2019; Patange, 2020). In harmony with different concrete requirements, many researchers have made efforts to reduce the utilization of cement and to maximize the performance (Kim et al., 2017), in the effort to explore the possible paths for the transition from a linear to a circular economy (Gnoni et al., 2017; Sparrevik et al., 2021). In recent years the use of substitute materials of concrete aggregate such as industrial waste and other aggregates has been of concern since these materials can be a solution in order to manage wastes from industry and also reduce environmental burden (Khan et al., 2020; Nursyamsi and Zebua, 2017; Sosoi et al., 2018). Migliore (2019) in his paper expressed the need to create virtual marketplace for wastes and by-products for exchange in concrete industry.

Tiwari et al. (2016) in their paper reviewed the feasibility of a wide variety of industrial by-products as fine aggregate replacement since concrete industry has acquired the credentials of being one of the largest consumers of some of the most vital natural resources. The cost of the concrete production has observed a sustained rise over a period of time and there has been an increasing shortage of fine aggregates, thus aggravating the situation further.

Some serious concerns have been raised repeatedly over the detrimental effects of reckless extraction of fine aggregate from river bed. An example of industry waste as replacement for fine aggregate is bottom ash from thermal power plants as presented in (Kumar Aswin et al., 2017; Malarvizhi and Mohanraj, 2016).

The overviewed literature indicated numerous papers on the possibility of applying waste LCD glass in concrete and related products. The possibility of LCD waste glass as sand replacement in concrete was highly researched as presented in (Rashad, 2015; 2014). Lin et al. (2009) have determined that thin film transistor LCD (TFT-LCD) waste glass can be regarded as a good glass– ceramic material. Similar research where waste LCD was replacement for fine aggregate was conducted by Góra et al., 2019. Their research showed that the waste obtained from LCD panels can be used as fine aggregate, e.g. at 1%, 2%, 3% sand replacement, and obtained concrete was ecologically acceptable.

Ruello et al. (2016) have in their paper valorised the possibility of using LCD scrap from indium recovery in mortars on a laboratory scale. Their research showed that scraps can be used to manufacture mortars that the regulation classifies as a CS II and CS III (compliant with a use for general works or as a plaster for indoor/outdoor). Recent study from (Lo et al., 2020) suggests geopolymer with 10%-40% LCD waste glass can partially replace metakaolin with favourable mechanical characteristics.

The above-mentioned papers do not include the use of waste LCDs after metal extraction as partial substitution for fine aggregate. In overviewed literature most researched element for extraction is indium (Amato et al., 2019; Amato and Beolchini, 2018; Cui et al., 2020), classified as critical raw material (European Commission, 2014). Other than indium, extraction of nickel, palladium, chromium, zinc, lead, aluminium, copper, arsenic is researched through methods and technologies that mainly include mechanical-physical separation, supercritical fluid, and vacuum metallurgy (Zhang and Xu, 2016).

Prior to concrete production, metal extraction process was carried out with leaching treatment of waste LCDs in *aqua regia*. This paper presents experimental research that gives insights into mechanical properties of a new green concrete where waste LCDs after metal extraction processes were used as fine aggregate replacements in defined percentages (1%, 5%, 10%). Properties of obtained concrete were compared with properties of a concrete prepared with the same percentages of waste LCDs prior to a metal extraction process. Emphasize was on demonstrating successful ‘loop closing’ in accordance with circular economy after metal extraction.

2. Material and methods

2.1. Preparation of modified concrete

For research purposes materials used were identical to those that are incorporated into the concrete products of real facility in Croatia. Fine (0-4 mm) and coarse aggregates (4-8 mm, 8-16 mm) obtained by exploitation and refining of construction sand and gravel were used as aggregates for concrete preparation. As binder was used only Portland cement, CEM II/A-S 42.5R. Specific gravity of this cement is 3.11, and Blaine surface area 3,900 cm^2/g. The chemical composition of used cement is given in Table 1.

Chemical analysis was performed in accordance with the norm HRN EN 196-2:2013. Water from water supply system was used. The same components were used in all concrete mixtures to avoid its influence on the properties of new concrete. Preparation of concrete mixtures, moulding and evaluation of its properties was carried out according to standard procedures described in HRN EN 12390.

In Table 2 are given fineness modulus and water absorption for fine and coarse aggregates and LCDs used in this experiment. Waste LCDs were obtained from authorized waste treatment facility for WEEE in Croatia and milled on particles up to 10 mm in size (Fig. 1). Grinding mill was VM/60 model, weight 400 kg, width 1280 mm, length 690 mm and height 1640 mm, with a working diameter of the rotor area of 160 mm and a power of 1.1 kW.

Table 1. Chemical composition of cement

Constituent	***Result (% mass)***
Loss of ignition	1.07
Corrected loss of ignition	1.38
Sulfuric anhydride, SO_3	2.99
Residue insoluble in HCI and Na_2CO_3	0.49
Residue insoluble in HCI and KOH	0.30
Sulphide, S^{2-}	0.13
Manganese oxide, MnO	0.32
Total silica, SiO_2	21.90
Iron-oxide, Fe_2O_3	2.49
Alumina, Al_2O_3	6.06
Lime, CaO	60.17
Magnesia, MgO	2.87
Chlorides, Cl^-	0.042
Soda, Na_2O	0.33
Potassium oxide, K_2O	0.74
Soda equivalent, Na_2O	0.82
Carbon dioxide, CO_2	0.40

Table 2. Finennes modulus and water absorption for aggregates and LCDs

Aggregate	***0-4***	***4-8***	***8-16***	***LCD***
Fineness modulus	2.88	5.91	5.76	4.24
Water absorption (%)	0.5	0.5	0.5	0

The representative sample was prepared after quartering to obtain necessary mass for research. Total of seven experimental mixtures was prepared with 0, 1, 5, 10% of fine aggregate replacement. Three experimental mixtures were obtained with chemically untreated LCDs (10mm), while other three experimental concrete mixtures were prepared with LCDs (10 mm) after treatment in *aqua regia (HCl: HNO_3 = 3:1)* in rate solid/liquid 1:4. Reaction was conducted in controlled ventilated space for 48 hours. After precipitation, the residues of treated LCDs were neutralized in a solution of sodium hydroxide and dried up to constant weight and used as an aggregate replacement in 1%, 5%, 10%. Control concrete mixture was prepared without fine aggregate replacement. In Fig. 2 it is shown chemical composition of used milled LCDs after treatment in *aqua regia (HCl: HNO_3 = 3:1)*. Elements of our interest were nickel (Ni), chromium (Cr), zinc (Zn), lead (Pb), aluminum (Al), copper (Cu), arsenic (As), cadmium (Cd), indium (In), according to literature overview.

(a) (b)

Fig. 1. (a) Obtained waste LCDs from authorized facility; (b) milled LCDs (10 mm)

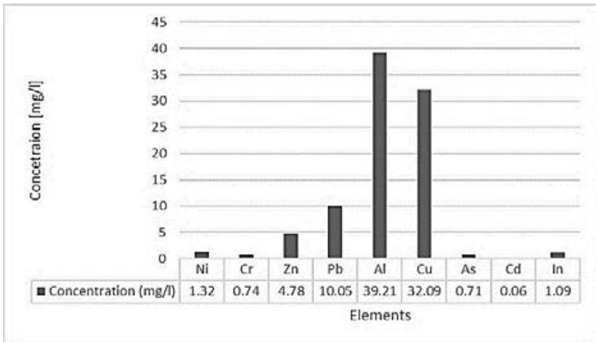

Fig. 2. Chemical composition of used LCDs after treatment in *aqua regia*

The recipe for concrete strength class C20/25 was used for preparation of testing samples. In the construction industry, given class of concrete can be used to make elements of load-bearing reinforced concrete structure, such as, for example, walls, slabs, beams and columns. In Table 3. is given working recipe for $1m^3$ of experimental mixtures of concrete class C20/25.

2.2. Properties of prepared mixtures

Determination of fresh concrete properties are of preventive character (Ukrainczyk, 1994) and in accordance were conducted for this research purpose. Following properties were tested: density, consistency and air content.

Table 3. Concrete mixture proportion per cubic meter for LCD and LCDt

Replacement wt.%	***Control Mixture***		***LCD 1%***		***LCDt 1%***		***LCD 5%***		***LCDt 5%***		***LCD 10%***		***LCDt 10%***	
Component	%	kg	%	kg	%	kg	%	kg	%	kg	%	kg	%	kg
LCD 10 mm	0	0	1	7	0	0	5	36	0	0	0	72	0	0
LCDt 10 mm	0	0	0	0	1	7	0	0	5	6	0	0	0	2
0 - 4	3.0	03	2.0	84	2.0	84	8.0	10	8.0	04	3	16	8.0	16
4-8	8.0	36	8.0	36	8.0	36	8.0	36	8.0	33	8	36	8.0	36
8-16	9.0	28	9.0	28	9.0	28	9.0	28	9.0	22	9	28	9.0	28
Cement 42.5 R		80		80		80		280		80		80		80
Water	0.6393	179	0.6393	179	0.6393	179	0.6393	179	0.6393	179	0.6393	179	0.6393	79
Dynamon LZF 35	0.3	0.84	0.3	0.84	0.3	0.84	0.3	0.84	0.3	0.84	0.3	0.84	0.3	0.84
Total		2327		2315		2316		2270		2256		2212		2213

Properties were tested according to standards described in HRN EN 12350 (Croatian Standards Institute, 2009a). Compressive strength and tensile splitting strength were tested according to standards described in HRN EN 12390 (Croatian Standards Institute, 2009b).

Apart from its strength, the concrete is also characterized by its workability. The workability is described as the ease with which concrete can be mixed, placed, consolidated and finished. The workability of concrete is measured by consistency which is tested in several ways. In this paper, the consistency test was tested by the slump test. The consistency is defined as the relative mobility, or ability of freshly mixed concrete to flow. It is indicative of the mix wetness. Usually, wetter mixes are more workable, with some exceptions to this rule. Accordingly, the slump height is the workability criterion of concrete (Konstantinidis, 2013), commonly known as a consistency.

In accordance with the standard HRN EN 12390-1: 2012 - Part 1: Shape, dimensions and other requirements for samples and molds, molds with dimensions of 150 mm (cube) were used. Thereafter, in accordance with HRN EN 12390-2: 2009 Testing hardened concrete - Part 2: Production and care of test specimens for strength testing, the inside of the mold was coated with a thin layer of non-reactive material to prevent adhesion of concrete to the mold and immediately after installation (distribution in molds) the concrete is compacted, in order to avoid segregation or the appearance of cement laitance. Thereafter, the test specimens were properly labeled, placed in a humid chamber at a temperature of 20 ± 2 °C and relative humidity ≥ 95%, left in the mold for 24 hours, then removed and left in the humid chamber until the test day, i.e. a total of 28 days.

One result of strength is the average value of three specimens' dimensions 150x150x150 mm after 28 days curing.

3. Results and discussion

3.1. Fresh concrete properties

Summarized representation of fresh concrete properties is given in Table 4. The density of fresh concrete was tested each time the consistency is measured. It is tested by incorporation and compaction in a container of known volume. Concretes with a smaller amount of cement paste (cement + water), i.e. a larger amount of aggregate have a higher density. Changes in the amount of air have the greatest impact on changes in the density of concrete, so by measuring the density of concrete, the amount of air in concrete can also be determined (Ukrainczyk, 1994).

When observing concrete mixtures with untreated LCDs a slight increase of fresh concrete density value was noticeable in regard to the LCD_t. According to the Herak-Marović, (2007), the density value of normal concrete is in range from 2000 to 2600 kgm^{-3}. Therefore, regardless to the percentage of untreated or treated LCDs incorporated in concrete composition, variations of measured density values were corresponded with the range found in literature.

An increase in air content value was noticed in LCD_t concrete mixtures in comparison with concrete mixtures with untreated LCD samples. The air content has an impact on the concrete strength, but in this research air content did not change considerably. Thereby, comparing the results it is noticeable that mixing the treated LCDs into the concrete increases the amount of pores filled with air, while mixing the untreated LCDs reduces the amount of pores filled with air. The deviation of granulometric curve of the aggregate from the optimum grain packing causes higher values of air content value, which is particularly noticeable for concrete with smaller amounts of cement paste (Ukrainczyk, 1994). This can also be observed in similar research described in (Melnjak et al., 2019; Premur et al., 2018).

Flowability is important fresh concrete property because it influences the construction quality. As a standard, in practices, slump is often used to evaluate the flowability (Guo et al., 2020). Guo et al. (2020) in their review paper noted inconsistent effects of glass on the slump in relation to the slump value of control concrete mixture. It is noticeably decreased for the concrete mixture where 1% of treated LCDs samples. A slight increase in the settlement of concrete with non-treated LCDs for 1 and 5% is noticeable, after which there is a significant decrease in concrete with 10% of non-treated LCDs. In LCD_t with 5% and 10% replacement, a significant increase in slump value was noticeable. Respectively, if the untreated LCDs samples are used as an aggregated substitute in concrete mixture, it should be up to 5% in order to preserve the concrete consistency.

Góra et al. (2019) report that the impact of LCD additive with significantly smaller amounts, i.e. 1%, 2%, 3%, and its mineralogical and chemical characteristics did not significantly affect the consistency of the mixtures. This was confirmed for concrete with untreated LCDs in this paper.

3.2. Results of hardened concrete mixtures

The most important property defined for hardened concrete is compressive strength (Bamforth et al., 2008). Strength tests were carried out following 28 days from sample molding as shown of Fig. 3 and curing as shown in Fig. 4. In comparison to the control mixture, the values of obtained mechanical properties (compressive and tensile splitting strength) can be seen in Table 5. The compressive strength of concrete is almost the same or increases slightly only for 1% replacement, both for treated and untreated LCD glass. The decrease of both concrete strengths is probably consequence of weaker surface binding of LCDs fragments with cement and poorer mechanical properties of LCD particles. Similar was observed by Góra et al. (2019) and Premur et al. (2018).

Table 4. Results of fresh concrete mixtures

Property	***Mixture***						
	LCD				***LCD_t***		
	0%	***1%***	***5%***	***10%***	***1%***	***5%***	***10%***
Density, kg/m^3	2329	2333	2337	2323	2306	2281	2292
Air content, %	3	2.4	2	1.8	3.3	2.8	3
Consistency, mm	150	160	160	110	90	180	210

LCD_t – concrete with LCD residues after treatment in aqua regia

a) b) c)

Fig. 3. Preparation of test specimens: a) standard molds; b) distribution in molds; c) test sprecimens after 28 days

Table 5. Results of mechanical properties of concrete

Property	***Mixture***						
	LCD				***LCD_t***		
	0%	***1%***	***5%***	***10%***	***1%***	***5%***	***10%***
compressive strength, MPa	33.6	33.4	28.6	21.6	35.53	30.67	27.4
st.dev.	0.75	0.37	0.11	0.45	0.90	0.57	0.85
tensile splitting strength, MPa	2.99	2.56	2.47	1.89	3.75	3.44	2.88
st.dev.	0.15	0.07	0.48	0.32	0.32	0.13	0.20

Fig. 4. Curing of concrete samples

The LCD concrete with 1% and 5% replacement are in accordance with the strength criteria for C20/25, while the replacement of aggregate with 10% of the same samples is not adequate. It can be noticed that LCD_t concrete with a partial substitution with 1% shows the higher compressive strength compared to the control concrete mixture. With higher replacement percentage tensile splitting strength decreased for concrete samples, but it is still inside the proscribed criteria for C20/25 (Bamforth et al., 2008). This result is similar to that obtained by (Her-Yung, 2009). Some researchers reported that the use of waste glass improved the properties of concrete, while other researchers reported opposite results. The inconsistent results of the effect of waste glass hinder the acceptance of glass in producing concrete, as presented in (Guo et al., 2020). Nevertheless, the concrete with the 5% and 10 % of replacement with the treated LCDs samples are in accordance with the strength criteria for C20/25.

In Fig. 5 are shown test samples after rupture, i.e. tensile splitting test, in order to show the broken surface. The concrete with partial substitution with 1% of treated LCDs samples showed higher values of concrete tensile splitting strength compared to the control concrete, while decreasing with higher replacement. This study indicated higher compressive and tensile strength values for concrete with LCD_t as opposed to concrete with untreated LCD. According to (Wang and Huang, 2010), the strength and durability of concrete is influenced by the proportion of the glass part of the LCD. Therefore, it can be concluded that the processing of LCD, i.e. indium extraction, improves the separation of glass particles from the LCD, which indicates higher values of concrete strength with treated LCDs. The tensile strength of concrete is many times less than the compressive strength (1/5 to 1/15).

Fig. 5. Test samples with LCD after tensile splitting test

It is tentatively assumed that the ratio of tensile and compressive strength of concrete is 1/10 (Herak-Marović, 2007). Because of high test variability of tensile testing, the compliance is based on the measurement of compressive strength (Bamforth et al., 2008).

4. Conclusions

Every year, more and more WEEE is generated in the world, and it is necessary to find a way how this waste can be adequately recycled without consequences for the environment and human health. According to some research, mixing waste LCDs in concrete, bricks, ceramic tiles, etc. is improving their characteristics and performance. In the experimental part of this paper, waste LCD panels milled to 10mm were used as a replacement for fine aggregate (sand) in concrete in different percentages (1%, 5%, 10%). Concrete was prepared with both milled waste LCD panels and residues of LCD after metal extraction in *aqua regia* to explore possibility of simple solution for this type of waste.

This research verified that the satisfactory results can be obtained both from a technical and environmental aspect, which is a significant scientific contribution to resource recycling and environmental protection since the need for natural sand could be reduced, but also residue after metal extraction could be managed as secondary raw material in accordance to circular economy. The results showed that with higher replacement the compressive and tensile strength of concrete decrease. Only the results for 1% and 5% replacement for both untreated and treated LCDs showed a similar compressive strength value as the control mixture (class C20/25). The compressive strength of control mixture (0% replacement) was 33.6 MPa, while for 1% replacement with untreated LCD the compressive strength was 33.4MPa as opposed to 35.53 MPa for concrete with same replacement with LCDt. For tensile splitting strength control mixture showed 2.99 MPa, while for 1% replacement with only milled untreated LCD this value was 2.56 MPa opposed to 3.75 MPa in concrete with treated LCDs in same replacement percentage.

It can be concluded that LCD processing enhances extraction of glass particles from the LCD as indicated by higher values of concrete strength with mixed treated concrete. The research presented in this paper justifies the usage of untreated and treated LCD for mixing into concrete, but only for 1% and 5% replacement in both cases.

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