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INFLUENCE OF USED WASTE CATHODE RAY TUBE GLASS ON ALKALI SILICATE REACTION AND MECHANICAL PROPERTIES OF MORTAR MIXTURES

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Abstract. *Rapid transition of electronic device manufacturing industry has led towards the increase of glass waste quantities, which are still being speculated. This resulted in increasing research on the use of waste glass in many different industries. In this study, the impact of using grounded waste cathode ray tube (CRT) glass as aggregate replacement (AR) on the alkali-silica reaction (ASR), mechanical properties and structure and microscopy of mortar were examined and reported. Crushed waste CRT aggregate was used to replace 0, 25, 50, 75 and 100% of natural limestone aggregate in mortar bars. ASR expansion values of mortar with added waste glass were investigated and tested for observation period according to Ultra-accelerated mortar-bar test. The results showed that the increase of AR percentage resulted in higher susceptibility to ASR. Mechanical properties and microscopy of mortar mixtures showed the potential of using waste CRT glass, due to the small difference between tested mixtures.*

Key words: *mortar, cathode ray tube glass, alkali-silicate reaction, mechanical properties, microscopy, aggregate replacement*

1. INTRODUCTION

In recent years, due to a greater demand for newer and more efficient products, the world has become a consumer-oriented society, which has ultimately led to the generation of various types of waste in huge quantities, some of them still being unknown [1]. Statistical reports prove that the European Union countries generate more than 33 Mt of different types of glass [2]. Due to rapid transition of screen manufacturing industry, there has been a rise in use of liquid crystal displays (LCD) in the form of electronic devices, but mostly in monitors

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and television sets. Such devices, which have more energy efficient displays than cathode ray tube glass (CRT), are also one of the leading environmental problems from the ecological point of view [3][4][5]. Since nowadays building and construction industry want to follow sustainability trends, glass is put to use with various applications whose justification is proven by several scientific studies in the field of building materials [6]. Some of the possible applications of waste glass in construction industry are in concrete paving blocks products, drainage application, roadway constructions, special concrete types, architectural applications, admixtures in brick manufacturing, foam glass, tableware glass, glass fibres, glazing ceramics. etc. [7][8]. Glass that is used in cathode tubes is based either on lead silicates (curved glass from the funnel coated with coating of lead and iron, which is hazardous waste) or aluminium silicate (in the form of curved glass that contains aluminium, titanium and vanadium coated by coatings of aluminium basis, and the flat screen glass powdered on by phosphate powder, composed of aluminum, vanadium, titanium, barium and lead to the limit of 0,085%, which is non-hazardous waste).

When it was discovered that waste glass might be used in construction industry as a building material, researchers also found out that glass is unstable in regard to alkali-silicate reaction (ASR) occurrence. Failures or deterioration of concrete structures, due to ASR, results from the reactivity within the concrete material, which leads to appearance of cracks on the surface and inside the volume of a hardened product. ASR is a phenomenon of concrete durability problem, although in most cases the aggregates used are chemically inert materials [9]. Several studies were carried out to examine possible ASR of reused waste glass in concrete and concrete products [1]. Monteiro P.J.M. et all. (1997) [10] used ASTM C 1260 method to measure the mortar bar specimen's expansion, microstructure and gel composition of mortar mixtures containing natural pouzzolan, fly-ash, and slag, made with two different types of Portland cement (PC). Yuksel et all. (2013) [7] investigated evaluation of three different tests (ASTM C1293, Rilem AAR-2, and microbar test method) for determining ASR reactivity of glass aggregate. Moncea et all. (2012) [11] investigated immobilization of waste CRT glass with high content of Pb and mechanical properties of mortar mixtures in three types of matrices Portland cement, slag cement and alkali activated slag binder. Tung-Chai Ling et al. (2013) [12] investigated the effects of particle size of CRT glass on properties of cement mortar made by using glass as sand replacement. Nirut Lairaksa et al. (2012) [13] reported possible use of CRT glass as a fine aggregate in a self-compacting. Hongjian Du. et all (2013) [14] investigated the influence of content, color and particle size of waste glass to ASR expansion of mortar. Also, they conducted a research on mortar mixtures where cement was replaced by mineral admixtures such as fly ash, silica fume, glass powder and ground granulated blast - furnace slag. D. Grdić et al. [15] investigated the change of properties of fresh and hardened cement mortar which occurred due to replacement of the aggregate with the CRT glass. Also, authors recommended that further research should be done on influence of ASR on mortar properties, shrinking due to drying, and modulus of elasticity. Rashad Alaa (2014) [2] studied fresh properties, mechanical properties, ASR, abrasion resistance, water absorption, etc. of mortar and concrete. In his study, mixtures containing waste glass as fine aggregate replacement have been tested.

For the purpose of determining ASR, several tests for ASR investigation can be used. During this research, it was found out that in most cases Rilem standards can be used to successfully identify the ASR reactivity of a tested material or mixture in short research period. The accelerated mortar bar test according to previous research seems to be a good precision test which tends to be the most effective [16].

In this study, the effect of CRT glass in mortar on ASR and mechanical properties was examined and reported. The ASR was investigated using standard Rilem TC 106-2 AAR Ultra-accelerated mortar-bar test (UAMBT) [17].

2. ALKALI AGGREGATE REACTION IN MORTAR

Alkali-aggregate reaction has two forms and they are: alkali-silica reaction (ASR) and alkali-carbonate reaction (ACR) [18]. ASR is a critical factor which determines the quantity of glass-sand content used in mixtures. While using waste glass in mortar or concrete, in the form of partial substitute of cement or aggregate, there is always a possible reaction in cementitious matrix or mortar [19]. Some aggregates have a possibility of reacting with alkali hydroxides in mortar or concrete, which has an impact on expansion and cracking of concrete in the service period [18]. In general, this phenomenon arises between alkali oxides, constitutes of cement, and aggregates that contain reactive silica foams [20]. In fact, when amorphous silica in glass is susceptible to attraction by the alkaline environment it would depolymerize to form a monomer $Si(OH)_4$. Further, monomer reacts with alkalis such as K+, Na+, and Ca+ to form ASR gel that can absorb water, induce inertial stress, and at the end induce severe cracking and damage [14]. In general, ASR is a phenomenon of reaction between the hydroxyl ions (OH−) in the pore solution and reactive silica-based ones on the aggregate. The alkalis initially contribute to the high concentration of hydroxyl ions in solution and formation of expansive alkali-silica gel. When poorly-crystalline hydrous silica is exposed to a strong alkaline solution, there is an acid-base reaction between the hydroxyl ions in solution and the acidic silanol (Si–OH) groups [21]. ASR causes expansion that takes place in two characteristic phases. The first phase is characterized by ASR gel showing, while in second, ASR gel combines with the moisture of the media, which causes expansion [20]. Previous research showed that using of low alkali cements might significantly reduce or completely eliminate this effect [19]. In general, ASR is a phenomenon in glass concrete or mortar that is still hard to predict before implementing competitive tests [14].

2.1. Materials and methods

Nowadays there is a real need for immediate detection of alkali reactivity of concrete made of alkali reactive aggregates. The potential alkali reactivity of aggregates comes forward in shape of mortar bar expansion [22]. Mortar bars consisting of alkali-reactive aggregates should be tested by rapid tests, far quicker than traditional alkali aggregate reactivity tests. Most common alkali reactivity detection tests are summarized in Tab. 1. Some of them are adopted as national standards for testing ASR in concrete products. On the other hand, one must have in mind that the national level regulations on ASR are usually developed based on local experience. Although it is expected that longer tests result in higher accuracy. Their main aim is to improve and optimize concrete mix design and flexibility of aggregate material, and incorporate aggregate properties the option of performance testing in their possible provisions [22][23].

Table 1 Alkali reactivity detection tests [9]

Name of the test	Procedure of using
Rilem TC 106-2 AAR Ultra-accelerated mortar-bar test (UAMBT)	In accordance to ASTM C 227 standard, mortar bars 25 mm x 25 mm x 285 mm or according to Note 1 40 mm x 40 mm x 160 mm in size should be prepared. After 24 hours prisms should be demolded and initial length should be measured. Prisms are then placed in water and heated in the oven to 80°C for the next 24 hours, initial length should be measured, zero expansion must be taken before specimens cooling. Prisms should be immersed into 1M NaOH solution already heated to 80°C. Specimen expansion must be observed periodically for 14 days. Mix design is recommended, so water/cement ratio is 0.47 by mass. Proportion of the dry materials is 1 part of cement to 2.25 part of the aggregates by mass. According to standard ISO 6274, a set of sieves should be settled to 4 mm, 2 mm, 1 mm, 500 µm, 250 µm and 125 µm [16].
ASTM C227 Mortar bar test	In this test, mortar bars should be placed into the heat-sealed. Courts for placing should be made of polyethylene, in the form of bags, and filled with approximately 10 ml of water. Expansion should be observed during period of 24 months. Expansion results are limited to 0.05% and 0.10% to the period of 3 and 6 months respectively. Late expansions of alkali-silicate reactive aggregates cannot be higher than 0.10% at 18-24 months period [24].
Danish salt method	Three ASTM mortar bars are exposed to NaCl solution that should be heated to 50°C. Expansion values are limited to 0.10% in the salt solution. Depending of aggregate, reports should be given on weeks 8 and 20 [9].
Chinese autoclave test	According to this method, mortar bars made of proportion material and mix design suggested in ASTM, with aggregate size 0,15 – 0,75 mm, are demolded after one day, cured at 100 °C stream curing for 4 hours, and later immersed into 10% KOH 150 °C heated solution for 6 hours [9][25].
NBRI method	This method is obtained by Oberholster and Davies. Mortar bars should be exposed to 1M NaOH solution at 80°C for the period of 14 days. Expansion should be measured in warm conditions. Expansions are limited to 0.10 to 0.25% [9].
Duncan method	Mortar bar standard ASTM dimensions are exposed to 100% relative humidity and to temperature. Heating temperature should be seated to 64°C for the observed period of 12, 16 and 26 weeks. Mortar expansion is limited to 0.05% [9][26].

3. EXPERIMENTAL PROGRAM

In order to assess the ASR reaction and mechanical properties of mortar bars, 40 mm x 40 mm x 160 mm bars were cast using Rilem TC 106-2 AAR Ultra accelerated mortar bar test (UAMBT). The reference mortar (E) comprised of cement and natural crushed limestone aggregate. Replacement ratios were selected as 25, 50, 75 and full replacement (100%) of aggregate with waste CRT glass (AR) by mass. These mixtures were designed as 25% (25 AR), 50% (50 AR), 75% (75 AR) and 100% (100 AR). The mixture proportions of mortar are shown in Table 2.

Table 2 Relative proportions of material used for preparation of mortar bar mixtures

Mixture name		E	25 AR	50 AR	75 AR	100 AR
w/c		0.47	0.47	0.47	0.47	0.47
cement		600	600	600	600	600
Mass retained between sieves (g)	Mineral aggregate					
	0.125-0.25 mm	202.5	151.87	101.25	50.63	0
	0.25-0.5 mm	337.5	253.12	168.75	84.38	0
	0.5-1 mm	337.5	253.12	168.75	84.38	0
	1 - 2 mm	337.5	253.12	168.75	84.38	0
	2 - 4 mm	135.0	101.25	67.50	33.75	0
	Waste CRT glass					
	0.125-0.25 mm	0	50.63	101.25	151.87	202.5
	0.25- 0.5 mm	0	84.38	168.75	253.12	337.5
	0.5- 1 mm	0	84.38	168.75	253.12	337.5
	1- 2 mm	0	84.38	168.75	253.12	337.5
	2- 4 mm	0	33.75	67.50	101.2	135.0

3.1. Materials

In this study, Portland cement, CEM I 52,5R [27] manufactured by CRH Popovac Serbia, with alkali content of 1.03% was used. The chemical composition of Portland cement is given it Table 3.

Table 3 Chemical composition of used cement CEM I 52,5R

Chemical element	%	Chemical element	%
LOi	2,26	MgO	2,2
IR	0,09	SO_3	3,05
SiO_2	19,3	K_2O	0,91
Al_2O_3	4,28	Na_2O	0,21
Fe_2O_3	2,87	P_2O_5	0,06
CaO	62,8	Cl	0,008

The purpose of the study was to compare the results of ASR and mechanical properties of mortar bars containing waste CRT aggregate with mortar containing crushed mineral aggregate. Crushed limestone aggregate used for making mortar bars was from Dolac quarry. The aggregate used was sieved with sieves according to standard, having square apertures of 4 mm, 2 mm, 1 mm, 500 µm, 250 µm and 125 µm, with maximum nominal size of 4 mm [28].

Used recycled panel glass of recycled CRT was granted by “Jugo-Impex” d.o.o. Niš company and crushed by fabric mill. By using “hot wire” method, screen glass was removed. This step was important due to different chemical composition of the glass. Taken from the device, glass was laboratory tested by removing harmful film from the outer and the inner tube of the CRT glass funnel. Several attempts of experiments were made. The coating of lead and iron on the outer side of the funnel was rinsed with regular water and removed in its entirety. Lead and iron based coating on the inner side of the funnel was hard to remove, except with mechanical procedures. Glass was immersed in 10% solution of nitric acid for 24 h. After the period of treatment, there has been no reaction leading to removal of the surface coating. The surface was treated with nitric acid in higher density, which has given no results. Aluminum based coating, on the outside was rinsed with water in the presence of abrasive paste. Aluminum based coating, on the inside, was submerged in 10% solution of hydro- chloric acid for 24 h. Reaction between aluminum and the acid was complete. Another possibility of removal is mechanical treatment of the surface in the form of scraping. Chemical composition of non-hazardous glass that is used without negative impact is given in Tab. 4. Used waste glass aggregate was sieved using sieves according to standard [28] non-hazardous CRT glass was sieved to sieves 4 mm, 2 mm, 1 mm, 500 µm, 250 µm and 125 µm.

No chemical admixtures were used in the experiment. Standard tap water was used during the mortar production in all mixtures.

Table 4 Chemical composition of used waste CRT glass

Chemical element	mg/kg	Chemical element	mg/kg
As	0,0	Pb	27,5
Be	0,2	Sb	9,5
Cd	0,2	Se	2,0
Co	10,3	Sn	5,7
Cr	6,0	Te	5,5
Cu	0,0	Tl	4,7
Mn	2,7	V	74,5
Ni	51,5	Zn	2100

4. RESULTS AND DISCUSSION

4.1. ASR expansion

According to RILEM TC 106-2 AAR standard that was used, the results obtained within 14 days of soaking observation period exhibited a potentially harmful expansion (Fig. 1). Measurements were taken at 2, 7, 10 and 14 days. The observed period showed turbulent reactions that had been expected as chemical reactions between cement, aggregate and water. Measurements showed shrinkage and ASR expansion processes in the observed specimens in various percentages using crushed CRT glass as a replacement for crushed limestone aggregate. For the observed period of 2 days, the lowest percentage of expansion is found in the mixture group 50 AR (0,04%), while the highest percentage of expansion is found in specimens from the group 100 AR (0,23%). When observing period was 7 days, all observed specimens, except from group E, show the expansion process. The highest percentage of expansion has been found in samples of group 75 AR (0,60%), while the smallest percentage of expansion has been found from the specimens in group 25 AR (0,21%). When observing period was 10 days, the lowest expansion belongs to mixture 25 AR (0,31%), while the highest percentage is shown in mixture 100 AR (0,82%). Finally, while observation period was 14 days, as expected, the highest and lowest percentage expansion belong to the samples of groups 100 AR (1,15%) and 25 AR (0,42%) respectively. It should be noted that ASR expansion of 25 AR was slightly lower than that of 100 AR, probably due to the lower amount of CRT glass. The results show that 25% replacement of natural crushed aggregate with CRT glass might be sufficient for controlling mortar bars. Expansion showed cracks visible on the surface of all specimens (Fig. 4) of 75 AR and 100 AR groups.

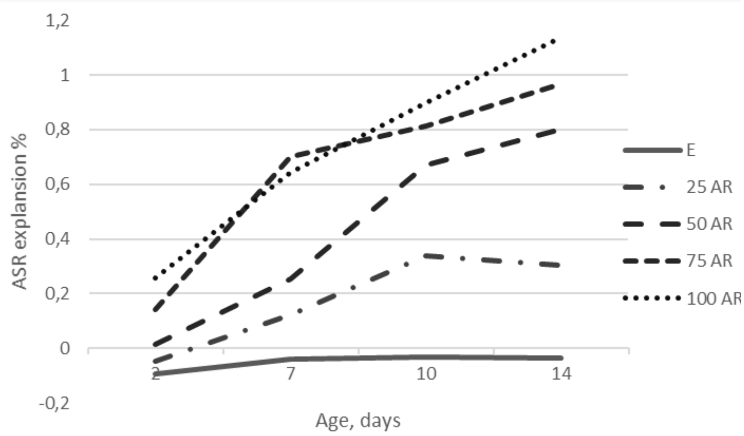


Fig. 1 Expansion of mortar bars containing waste CRT glass.

Shrinking is characteristic for reference (E) specimens and specimens where the crushed limestone aggregate was replaced by crushed CRT aggregate in the amount of 25% (25 AR) for a measurement on 2 days. Measured values for reference show that the greatest shrinking values were at the beginning of the observation period, then they show rapidly lower values that were constant during other measures. At 2, 7, 10 and 14 days, the measures were 0,086 %, 0,038%, 0,02 % and 0,03% respectively. The measured value for 25 AR that records shrinking at 2 days was 0,04%. Other values of mixture 25 AR show expansion. Recorded values at 7 days were 0,21%, and they increased by approximately 50% respectively at each subsequent measurement step. Other curves that represent the behavior of samples during the observed period show only expansion.

4.2. Mechanical properties of mortar bars

The compressive and tensile strength of 14 days old specimens with different replacements of natural aggregate with waste CRT glass are shown in Fig. 2. and Fig. 3. Considering that compressive and tensile strength of mortar bars is the most important mechanical property of the mixture, the results show satisfactory initial mechanical characteristics compared to control mixture E.

The results of compressive strength show expected reduction which corresponds to this percentage of natural aggregate replacement by waste CRT glass.

The measured strength values show maximum strength that fits to reference E (60,04 N/mm^2). Mixture 25 AR shows value 56,99 N/mm^2, that is for 5,1% reduced value than E. Mixture 50 AR shows compressive strength value reduced for 2,56% in comparison to 25 AR, which is for 7,6% lower strength than in mixture E. Measured values of 75 AG and 100 AR are 49,31 N/mm^2 and 42,23 N/mm^2 respectively. Difference in percentage of compressive strength values between E and 100 AR are 29,9%. Compressive strength diagram shows the expected trend of declining strength, which corresponds to the percentage of aggregate replacement by waste CRT glass. Percentage of compressive strength of mixture 50 AR is higher than arithmetic mean of the mixtures E and 100 AR, which proves a small difference of compressive strength reduction of mixture 25 AR and 50 AR.

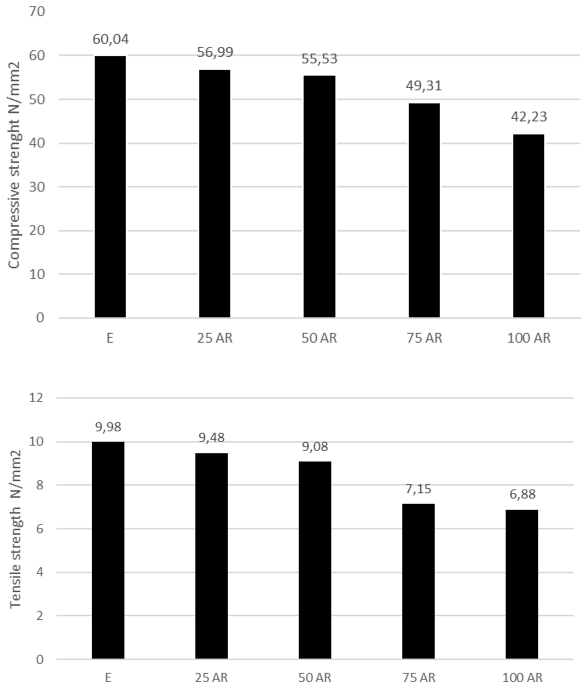
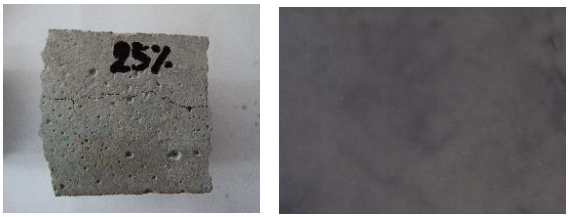


Fig. 2 and 3 Compressive and tensile strength of mortar bar with added waste CRT glass after 14 days.

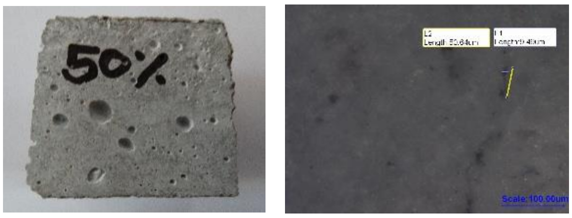
The results of tensile strength show reduction, like in compressive strength diagram which also corresponds to percentage of aggregate replacement by waste CRT glass.

25 AR - No crack observed



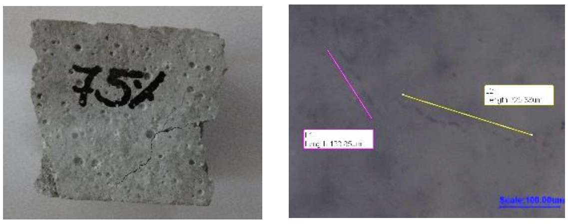
50 AR - Short crack observed

Width: $L1_{50\ AR}$=9.49 µm; Length: $L2_{50\ AR}$ =50.64 µm



75 AR - Crack observed

Length: $L1_{75\ AR}$ =133.82 µm; Length: $L2_{75\ AR}$ =225.38 µm



100 AR - Crack observed

Width: $L1_{100\ AR}$ =77.72 µm; Area: $P1_{100\ AR}$ =28176 µmSq;
Area: $P2_{100\ AR}$ =20513µmSq

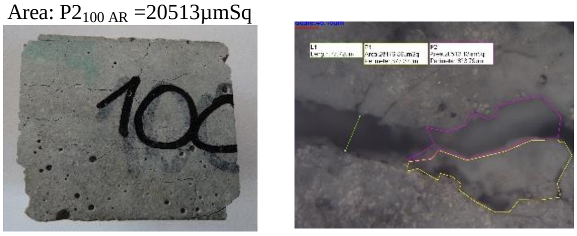


Fig. 4 Microscopy of AR mortar bars

The measured strength values show maximum strength of mixture E (9,98 N/mm^2). Mixtures 25 AR, 50 AR, 75 AR and 100 AR show tensile strength measured values 9,48 N/mm^2, 9,08 N/mm^2, 7,15 N/mm^2, 6,88 N/mm^2 respectively. Mixture 25 AR shows 5% lower tensile strength values than control mixture. Mixture named by 50 AR shows 4,2 % reduced values than mixture named by 25 AR, and 9% reduced values than mixture E. Like with compressive strength diagram, in the Fig. 3. that shows tensile strength, one can notice the downward trend in strength, which corresponds to the percentage of aggregate replacement by waste CRT glass. Values of compressive strength of mixture 50 AR are above arithmetic mean of E and 100 AR mixture values.

The photos of microscopy observed specimens are shown in Fig. 4. Cracks are visible on surface mixtures designated as 50 AR, 70 AR and 100 AR. Mixtures designated as 25 AR did not have microscopy surface visible cracks. By using microscopy, characteristic crack values were obtained on the samples surface. The measured values L150 AR and L250 AR of mixture 50 AR presents values of width 9,49 µm and length 50,64 µm, respectively, are visible on the samples surface. The measured cracks values named as L175 AR and L275 AR of mixture 75 AR presents length values 133,82 µm and 225,38 µm respectively. Measured crack L275 AR shows 4,45 times longer crack length value than L250 AR. Mixture 100 AR have characteristic crack width L1100 AR (77,72 µm), and areas P1100 AR (28176 µmSq) and P2100 AR (20513 µmSq). Measured L1100 AR width crack value is 8,2 times longer than measured width of L150 AR.

5. CONCLUSION

In accordance with test duration standard, observation of mortar shrinkage and expansion processes, led to the conclusion that during the observation period on ASR had a noticeable impact on the expansion of mortar with ground CRT glass used to replace a crushed limestone aggregate. The impact of ASR on expansion increased with higher percentage of recycled CRT glass used to replace crushed limestone aggregate. Expansion had a noticeable impact on the surface cracks. Reduced compressive and tensile strength values were attributed to significantly smoother surface of the CRT glass aggregates, as well as the fact that recycled glass aggregate had a harmful coating that prevented proper adhesion of the recycled glass aggregate’s grains and cement matrix. Aside from being prescribed by the RILEM TC 106-2 AAR guidelines used in this research, 14 days testing period is significant for the fact that the most important chemical reactions in the mixture occur during that time period. Mechanical properties of mortar mixtures showed the potential of using waste CRT glass, due to the small difference between strength of tested mixtures.

Further research should be conducted using the same testing methodology and guidelines using cements that contain mineral additives and mineral supplements as partially substitute of cement or aggregate. That might be in order to further reduce effects on ASR and improve mechanical properties of mortar bars and concrete.

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ISPITIVANJE UTICAJA KATODNOG STAKLA NA POJAVU ALKALNO SILIKATNE REAKCIJE I MEHANIČKE KARAKTERISTIKE MALTERSKIH MEŠAVINA

Razvoj industrije elektronskih uređaja poslednjih godina doveo je do znatnog povećanja nastalog staklenog otpada, čije količine još uvek nisu u potpunosti poznate. To je rezultiralo razvojem istraživanja koja se često baziraju na upotrebi otpadnog stakla u raznim proizvođačkim industrijama. U ovom radu ispitivan je uticaj otpadnog mlevenog stakla katodnih cevi (CRT staklo) kao delimične i potpune zamene agregata (AR) u malterskim mešavinama i uticaj takvog agregata na pojavu alkalno silikatne reakcije (ASR), mehaničke karakteristike, strukturu i mikroskopiju ispitivanih mešavina. Mleveno CRT staklo korišćeno je kao zamena prirodnog agregata u procentualnoj zastupljenosti od 0, 25, 50, 75 i 100%. Količina nastale ASR reakcije na uzorcima malterskih prizmi je ispitivana i testirana prema Ultra-accelerated mortar-bar test-u. Rezultati ispitivanja pokazuju da sa procetualnim povećanjem AR dolazi do pojave većeg uticaja ASR. Mehaničke karakteristike i mikroskopija malterksih mešavina u konačnom pokazuju veliki potencijal korišćenja CRT stakla.

Ključne reči: *malter, staklo katodnih cevi, alkalno-silikatna reakcija, mehaničke karakteristike, mikroskopija, zamena agregata*.