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Use and assessment of "e-plastics" as recycled aggregates in cement mortar

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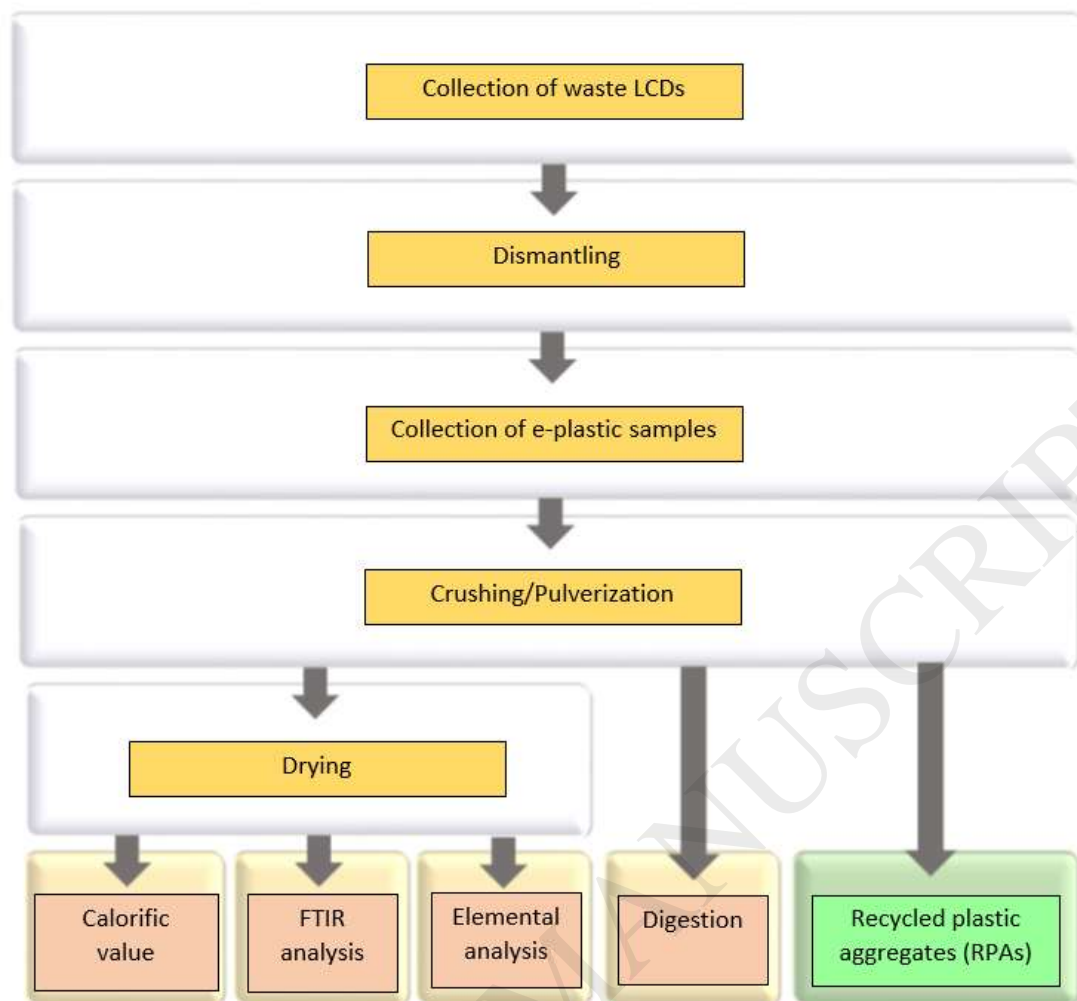College of Arts and Sciences, Center for Sustainable Development, Qatar University, P.O. Box: 2713, Doha, Qatar. Tel.: +97444037559

Graphical abstract

Highlights

- E-Plastics were recovered from waste liquid display screens (LCDs).
- The obtained e-plastics were compositionally and elementally analysed.
- ABS was partially used and assessed as a recycled aggregate (RA) in cement mortar.
- The effect of ABS in the properties of cement mortar was investigated.
- The 7.5% and 10% replacement by e-plastics favoured the strength of the specimens.

Abstract

In this study we investigated the physical and mechanical properties of cement mortars, partially replaced with plastic (recycled plastic aggregate, RPA) recovered from WEEE (namely, "e-plastics"). The plastic housing of 14 LCD screens was sampled and, subsequently, compositionally and elementally analysed. Acrylonitrile-butadiene-styrene (ABS), being the most commonly found polymer in WEEE, was used as aggregate in the cement mortar. The replacement percentages (RPs) used were 2.5%, 5%, 7.5%, 10% and 12.5%, while the water to cement (w/c) ratio was maintained constant at 0.5 in all tests. The basic properties (e.g. compressive strength (CS), modulus of elasticity (MoE), density (ρ), porosity ($\varPhi$) and water absorption (WA)) of the created specimens were investigated. The obtained results, for all RPs, (especially those of 7.5% and 10%) exhibited an increase in the CS of the specimens by 15.4% and 7.8%, respectively, with the MoE decreased in both cases by 18.1% and 23.8%, respectively. The rest of the examined specimens' physical properties measured (ρ, $\varPhi$, and WA) indicated, also, greater strength but lower ductility than the reference (standard) specimen. Concluding, the use of recovered plastics from WEEE as recycled aggregates (RAs) in cement could potentially be proved a useful downcycling alternative for waste plastics.

3

ABBREVIATIONS

ABS	acrylonitrile-butadiene-styrene
BFRs	brominated flame retardants
CS	Compressive strength
CV	Calorific value
EA	Elemental analysis
EU	European Union
FRs	flame retardants
HDPE	High-density polyethylene
HIPS	high-impact polystyrene
LCD	liquid-crystal-display
MoE	Modulus of elasticity
ρ	density
PC	polycarbonate
Φ	Porosity
RPAs	Recycled plastic aggregates
PTMs	Potentially toxic metals
PVC	polyvinyl chloride
SoC	Substances of concern
WA	Water absorption
RAs	Recycled aggregates
WEEE	Waste electrical and electronic equipment
PET	Polyethylene terephthalate

1. Introduction

Owing to their complex and multi-elemental composition, waste electrical and electronic equipment (WEEE) (also known as "e-waste") is one of the most important waste streams that is expected to exceed 50 Mt within the year 2019 [1, 2]. Ferrous is the most prevalent element (ca. 50%) in WEEE, followed by plastics (with or without the presence of brominated flame retardants (BFRs) embedded in them) that account for ca. 20%, while copper, glass and aluminum are found in respective individual percentages of <10% (see Fig. 1a) [3, 4].

The plastics fraction present in e-waste ("e-plastics"), and especially in computer screens, is consisted of different types of polymers [5] (see Fig. 1b), mostly thermoplastics (e.g. acrylonitrile-butadiene-styrene (ABS), mixture of ABS and polycarbonate (ABS/PC) and high-impact polystyrene (HIPS)), whose major advantage is the reversibility of their formation and, thus, their potential of multiple reuse [6, 7]. In a liquid crystal display (LCD) screen, for instance, 25% of its composition is plastics [8, 9].

Nonetheless, the appropriate recovery and recycling of the e-plastics fraction is a quite challenging task, mainly due to the variety of polymer types and the releasing potential of harmful gases from any existing BFRs [10]. The current recycling routes for e-plastics include mechanical recycling, chemical recycling with energy recovery, as well as their use in the construction sector as recycled aggregates (RAs) [11, 12].

The chemical stability of thermoplastics, when present in alkaline solutions like the ones found in concrete, constitute one of the major reasons for continuous research into that area [13]. According to the results obtained from several studies, plastic recycled plastic aggregates (RPAs), though may not always enhance the mechanical properties of the specimens tested, could extend their lifetime and their

5

thermo-insulate properties [14-16]. Some of RPAs' reported advantages include: a) flexibility and adjustability, b) lightweight nature, c) resistance and durability in chemicals, d) thermos and electrical insulate properties and e) low production cost. On the other hand, poor cohesive properties leading to reduced compressive strength (CS), modulus of elasticity (MoE) and overall concrete durability, as well as a relatively low melting point constitute some of their reported disadvantages [13].

Subramani and Pugal [17] reported that a 10% plastic replacement in concrete results in higher durability, whereas increasing the replacement ratio up to 20% can produce satisfactory and acceptable specimens [17].

Manjunath [13] on the other hand, reported a decrease in the CS and durability of concrete enhanced with RPAs, when increasing the plastic RP from 10%, to 20%, to 30%, under a constant w/c ratio of 0.5; still, though, obtained properties were within operational and legislative limits [13]. This could most possibly be attributed to the hydrophobic nature of plastic and to the reduced cohesion between materials.

Similarly, reduced compressive and tensile strengths were observed by the use of recovered high-density polyethylene (HDPE) from e-waste in concrete, regardless of the w/c ratio used [18, 19].

One of the most commonly used RPA is polyethylene terephthalate (PET). Silva et. al. [20] investigated the replacement of 7.5% coarse aggregates with PET. The results indicated a 5.1% reduction in the CS of specimens, but within permissible limits [20].

In another study, a replacement of up to 20% with PET (maintaining the w/c ratio at very low levels) produced satisfactory and encouraging results in the use of RPA in the construction sector [21].

Moreover, though the use of recovered ABS and polyvinyl chloride (PVC) from waste photovoltaic panels as RPA in 5% and 10% resulted in specimens with lowered

6

strengths, the researchers concluded that the addition of ABS could potentially enhance the insulating properties of new specimens [22].

In general, during the last decades several studies have focused on evaluating and assessing the use of recovered plastics (originated or not from e-waste) in concrete, and some of them have also critically and comprehensively reviewed their use and effects [11, 14, 23-25].

The main aims and objectives of the present study are: a) to investigate the use and effects of e-plastics (recovered from waste LCDs) when used as RAs in concrete, b) to determine the composition and characteristics of the retrieved e-plastics and c) to evaluate the effects that the partial replacement with RPAs has on the mechanical and physical properties of concrete.

2. Material and methods

Waste LCDs were collected from the Inter-Municipal Solid Waste Management Company of Chania Prefecture, Crete, Greece. After a thorough dismantling, e-plastics were recovered, either from the front or the back cover of the LCD screens, and were further dried and cut down to smaller pieces and/or fibers, using graters and scissors, to further facilitate their subsequent physicochemical characterization and analysis, as shown in Fig. 2. The obtained samples were, in turn, classified in three categories (namely ABS, ABS/PC, ABS/HIPS and ABS/PC/HIPS) according to the polymer type contained in them, as shown in Table 1.

The calorific value (CV) was measured using an AC-350 by LECO, whereas for the determination of the toxic metal content, the samples were first digested in a MARS 6 Microwave Reactor System CEM, following EPA method 3051, with subsequent analysis of the digestates in an ICP-MS (Inductive Coupled Plasma Mass

7

Spectrometer) 7500cx by Agilent Technologies. The Fourier-transform infrared spectroscopy (FTIR) analysis was performed using a Perkin – Elmer system 1000 and the elemental analysis via the use of a EuroVector, Elemental Analysis CHNS-O.

In turn, e-plastic samples sized < 4mm and contained only ABS were selected to partially replace sand in the manufacturing and study of cement mortar specimens. The cement used was CEM II/A-m 42.5 N type and the sand was limestone type sieved at <4mm and priory dried at 105 °C for 24h. The specimens were prepared according to ASTM C 109/C 109M – 07 method (in 5 x 5 x 5 cubes, using the reccomended quantities of concrete, sand, water and e-plastic described therein) and the w/c ratio was kept constant at 0.5. Sand replacement by ABS was performed at 2.5%, 5%, 7.5%, 10% and 12.5% percentages. Specimens were left to mature for 7 and 28 days, respectively, since at the 28th day the specimen obtains its maximum strength. For each one of the five aforementioned replacement percentages (RPs), 3 specimens were analysed during the 7th and 28th day of maturation.

Finally, all prepared specimens were analysed in their mechanical (CS, MoE) and physical (density (ρ), porosity (Φ) and water absorption (WA)) properties.

3. Results and discussion

3.1. Calorific value (CV) and elemental analysis (EA)

CV and EA results, in comparison to several other studies in literature [10, 26-31], are presented in Table 2.

As it can be seen, all specimens displayed similar CV values, at ca. 39 MJ/kg, with the exception of one ABS sample that demonstrated an approx. 34 MJ/kg and two ABS/PC+HIPS specimens with values at around 41 MJ/kg. These results are in consistency with the limited existing literature, where RPAs have been reported to

8

exhibit similarly elevated CVs (close to the 43 MJ/kg of petroleum), mainly due to the high hydrocarbon content of plastics [32, 33]. In contrast, carbon and wood have much lower CVs, 30 and 19 MJ/kg, respectively [34].

With regards to EA of the samples, since plastics are mostly composed of hydrocarbons, carbon content is expected to be high. Any fluctuations in C, N and H percentages are mostly due to different manufacturing processes followed by each individual company.

More specifically, ABS is the only polymer exhibiting nitrile groups, whereas PC and HIPS contain a small amount of N_2, which most of the times is untraceable [31]. Thus, the sample with the code name LCD_2 (being an ABS) bears the maximum N_2 content (5.72%), whereas in the rest of the samples this percentage is decreased due to the presence of other polymers (see Table 2).

3.2 Concentrations of potentially toxic metals (PTMs) in e-plastics

The European Union (EU) initially established two Directives, 2002/96/EU and 2002/95/EU, along with their most recent recasts (2012/19/EC; 2011/65/EC) where certain limits for various substances of concern (SoC), among which various PTMs such as Cr, Cd, Hg, Pb, Br, are included; recovery and recycling targets for WEEE, are also regulated, therein [35-38].

The concentrations of several PTMs found in the e-plastic samples of this study are presented in Fig. 3a, whereas Figs 3b and 3c depict the respective range of their values. It is apparent that Si, Mg and Al are the most abundant metals in all samples (even if present in various ratios within the same type of plastic), indicating the presence of FRs [39, 40]. Exception to this was noted for sample LCD_12 (ABS/PC+HIPS), where Sn was measured at >30 ppm. In addition, Sb (mostly used in combination with Br in FRs) was found in <10 ppm and only in two samples (LCD_3

9

and LCD_5). Finally, Ti, Zn and Fe were also measured in considerable amounts [10, 41]. It is also noteworthy that Pb and Cr were found in traces, whereas Cd was practically undetected. Finally, Ca was measured in concentrations >100 ppm in almost all samples, which is due to the use of $CaCO_3$ as filler in most of the e-plastic samples [40, 42].

As a general observation it can be stated that concentrations of PTMs are much lower than the permissible limits and in all samples Mg and Al FRs are present.

3.3 FTIR analysis

FTIR diagrams for all samples (per polymer type) are shown in Fig. 4 (a, b, c and d).

Relative peaks at 2238 cm^{-1}and 968-912 cm^{-1} (Fig. 4a) indicate the presence of ABS, where the C≡N bond and the butadiene can be detected [10]. Since ABS and HIPS possess the same bonds, with 2 similar monomers (butadiene and styrene), nitrile is the only way to distinguish between them [31].

Absorption at 1494 και 1454 cm^{-1} indicates the presence of styrene, whereas at 762 και 702 cm^{-1} one can identify the bend of the benzoic ring. Finally the stretch between the double and single bonds (C = C, C – H) is shown by the peaks at 1602 cm^{-1} και 2924 – 2850 cm^{-1} [43]

With regards to PC, literature reports an identification peak at 1720 cm^{-1}. However, none of the examined sample presented such a peak, fact which indicates either the lack or the miniscule amount of this polymer type (Figs 4b and 4c).

Through FTIR analysis the presence of several oxides (mentioned in the previous section of this work) can also be identified (i.e. MgO, TiO_2 and SiO_2, at 2924 cm^{-1}, Al_2O_3 and Fe_2O_3 at 1366 cm^{-1} and $CaCO_3$ at 848 cm^{-1}) since they were abundant in all examined samples.

10

3.4 Application of RPAs in cement mortars

Several cement specimens (partially replaced with ABS in 2.5%, 5%, 7.5%, 10% και 12.5%) and a w/c=0.5 were prepared in order to test their response and changes in their mechanical and physical properties compared to the standard (control) sample (specimen). More specifically, CS, MoE, $Φ$, ρ and WA were investigated in the RPA modified specimens. The materials used for the preparation of the specimens are: a) ABS particles, b) cement type CEM II/A-m 42.5 N, c) properly sieved limestone sand and d) water.

3.4.1 Compressive strength (CS)

The CS results for all examined RPs and for both 7 and 28 days of specimens' maturation are shown in Figs. 5a and 5b.

As it can be seen, CS increases with maturation time regardless of RPs. This can be explained since under humid environment cement hydration is increased, therefore mortar cohesion is favoured [15]. A heterogeneous behaviour was observed since samples with 2.5%, 5% and 12.5% RPs exhibited a lower CS than the standard, whereas higher CS was measured for the samples with 7.5% and 10% RPs. More specifically, for 7 days of maturation a decrease of 13.4%, 17.3% and 23% in the CS is observed, for respective RPs of 2.5%, 5% and 12.5%. On the other hand, specimens with 7.5% and 10% RPs demonstrated a 26% and 33.4% increase, respectively. Similarly, on the 28 days of maturation specimens with 2.5%, 5% and 12.5% ABS replacement ratios showed a decrease of 13.2%, 28.8% και 42.3% in the CS, whereas the ones with 7.5% και 10% exhibited a 15.4% και 7.8% increment in CS.

The aforementioned results are quite interesting since an increased CS in RPAs is not a common phenomenon observed. However, there are a few studies in literature

11

that report similar and/or close findings to ones of this study [44, 45]. It is imperative to understand that CS is a multi-dependent property affected by a variety of parameters (e.g. w/c ratio, type and shape of RPAs, maturation temperature, etc.). Even the grain size of the sand or plastic used can affect the properties of the specimens.

On the other hand, a decreased CS in RPAs is a behaviour commonly reported in literature, mostly attributed to the hydrophobic nature of plastics that hinders the necessary hydration of specimens during maturation [11, 14]. In most cases, a CS decrease becomes more intense with increased replacements ratios.

3.4.2 Modulus of elasticity (MoE)

The MoE is highly dependent on the type of RPAs used in the specimens, since any deformation observed is strongly affected by the elastic deformation occurring tin the RPAs [11]. Other factors affecting the MoE are the w/c ratio, the Φ of the RPAs, as well as the RPs.

In general, as shown in Fig. 6a, MoE tends to decrease with an increase in the replacement ratio. This can be mainly attributed to the low elasticity of plastic and to the decreased density [46, 47]. The fact that specimens with 7.5% and 10% RPs exhibited high CS and MoE values, close to the ones of the standard, could justify their use as external coatings.

3.4.3 Porosity (Φ), density (ρ) and water absorption (WA)

These properties were measured in all 28 days maturation specimens, including the standard.

The porosity (Φ) is strongly affects both strength and WA of the specimen. It is usually dependent on w/c ratio and quality of RPAs. In practice low Φ values

12

correspond to appropriate distribution and connection of the RPAs into the cement mortar, thereby enhancing its natural properties. The Φ results of the present study are shown in Fig. 6b. As it was expected, taking also into account the CS results, in the 2.5%, 5% and 12.5% RPs, Φ was found in elevated values, whereas for the 7.5% and 10% specimens (where CS was reported at high values) Φ was decreased. In general, the addition of RPAs in cement leads to a decrease in Φ, due to their inappropriate distribution in the mortar and the hydrophobic nature of plastics [23].

WA is a cement property reflective of its volume and linkage to the permeable pores [14]. In several studies where a RPAs were partially used, WA was increased since the mixture of plastics and sand was not fully embedded in the cement mortar, thus creating greater gaps amongst the different materials [11, 20, 23]

In the present study, 2.5%, 5% και 12.5% replacement ratios resulted in an increase in the gap between RPAs and cement mortar (higher WA than the standard), whereas in the 7.5% and 10% specimens WA was measured at lower values than the standard (see Fig 6c). This trend in consistent with Φ and CS results since all these properties are interrelated and constitute an indication of the suitability of the specimen to be used or not in construction applications.

Finally, plastic is a lightweight material which, when added in cement mortar, it directly affects its density. ABS has a $\rho=1.0\text{-}1.4$ g/cm^3. According to literature, RPAs tend to lower the ρ of specimens [11], and in turn their mechanical properties.

In the present study, the density of the specimens exhibited small fluctuations for all RPs used (see Fig. 6d). In particular, for 2.5%, 5% and 12.5% replacement ratios, ρ appears lower than the standard, whereas in the specimens with 7.5% and 10% RPs ρ measured was slightly higher. This trend is in agreement with the CS results, appearing, also, higher in the respective replacement ratios.

13

The results of the present study indicate a rather bizarre behaviour of the RPA specimens. CS is only increased in the 7.5% and 10% RPs, even though the other RPs displayed CS values close to the ones of the standard. WA, Φ and ρ values were in agreement with the CS trend. It can be stated that the granulometry of sand and plastics used should have acted as a crucial factor to this behaviour, since the rest of the experimental parameters were kept constant at all tests performed. Although it was not determined which exact fraction of the sand was replaced by plastics (which were just mixed with sand in a 4 < ABS < 2 mm particle size), it is possible that during the preparation of the 7.5% and 10% RPs specimens, the best possible mixture result was achieved.

4. Conclusions

In the present study, the use and assessment of e-plastics as aggregates in cement mortar was investigated. In order to evaluate the RPA specimens, various RPs and their effect in the mechanical and physical properties of the created specimens were examined.

ABS was initially analysed and results indicated a high CV, lightweight material, suitable for use as aggregate in the construction sector. Its metal content was found within the permissible limits set by the EC, whereas the measurement of other metals i.e. Si, Mg and Al were indicative of the presence of respective FRs. The amount of Ca detected is also directly affiliated with the use of $CaCO_3$ in the plastics manufacturing. FTIR analysis also revealed the full composition of the ABS content and confirmed the aforementioned presence of the metals/metal oxides.

In turn, several RPAs were prepared using various RPs. The CS results were unevenly distributed demonstrating a –consistent with literature- decrease in CS of the

14

specimens, for 2.5%, 5% and 12.5% RPs, and an increase for 7.5% and 10% RPs; a fact that could mostly be attributed to the effective granulometry distribution of plastic in the structure of the cement mortar.

In contrast, MoE after 28 days of maturation exhibited a decrease indicating that despite the acquired strength of the specimens they became less elastic. This conclusion was reinforced by the results obtained for Φ, WA and ρ. The fluctuation of the latter properties was in consistence with the measured CS of the specimens.

Major parameters that can potentially affect CS are the w/c ratio and the granulometry (particle size) of the aggregate. In this study, w/c was maintained constant at 0.5; however, granulometry varied, a fact which may be responsible for the fluctuations in the results.

Concluding, ABS can be used in small quantities as a RPA, to produce external coatings, thereby serving as a valuable recyclable material. Similar applications include the use of PET in industrial scale as a RPA used in cement or in manufacturing roads.

Although literature is limited in the research and use of ABS as a RPA (most studies deal with PET) as a future work the authors would recommend a more thorough (e.g. microscopic) investigation of the specimens or the effect of other parameters involved (e.g. UV, cooling cycles, carbonation, etc.). The investigation of more RPs (especially within the range of 5%-10%) could shed further light on the observed increased CS of the specimens. Lastly, a chemical pre-treatment that would smooth the hydrophobic nature of plastics could potentially result to a better cohesiveness of the mixture, thereby enhancing the final specimen's properties.

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15

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List of Figures

Fig. 1. a) Typical WEEE composition (adopted by Ma et al. [3]) and b) distribution of polymer types within the WEEE plastics fraction (adopted by Yang et. al., [5])

Fig. 2. E-plastics recovered from waste LCDs in a) full, b) grinded and c) ready for drying sizes.

Fig. 3. a) Concentrations of PTMs in the examined e-plastic samples along with their respective ranges observed (b and c).

Fig. 4. FTIR spectra for all e-plastic samples analysed, per polymer type: a) ABS, b) ABS/HIPS, c) ABS/PC + HIPS and d) ABS/PC.

Fig. 5. Compressive strength (CS) of specimens for all examined replacement percentages (RPs) and for both 7 and 28 days of maturation.

Fig. 6. Obtained results for all examined replacement percentages (RPs) of specimens with regards to: a) modulus of elasticity (MoE), b) porosity (Φ), c) water absorption (WA) and d) density (ρ).
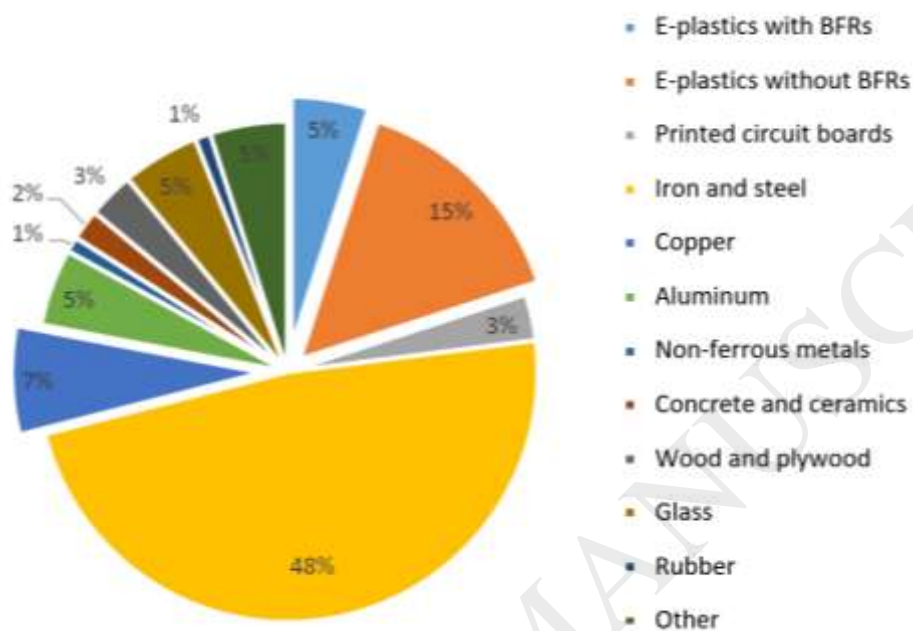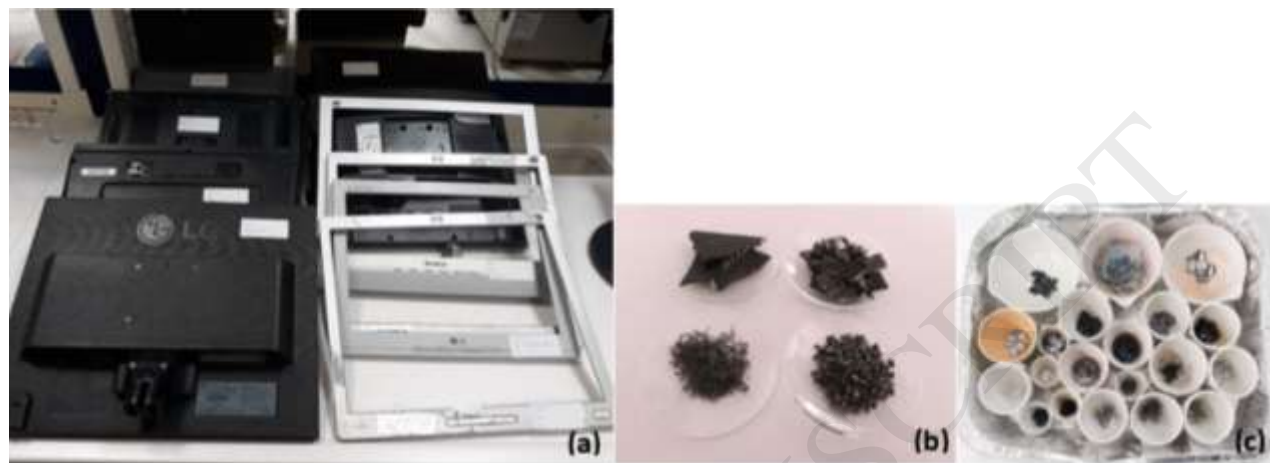
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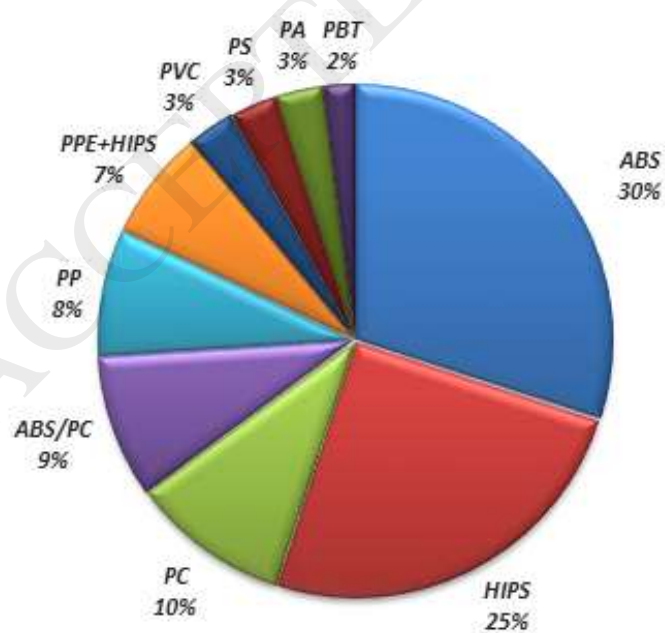
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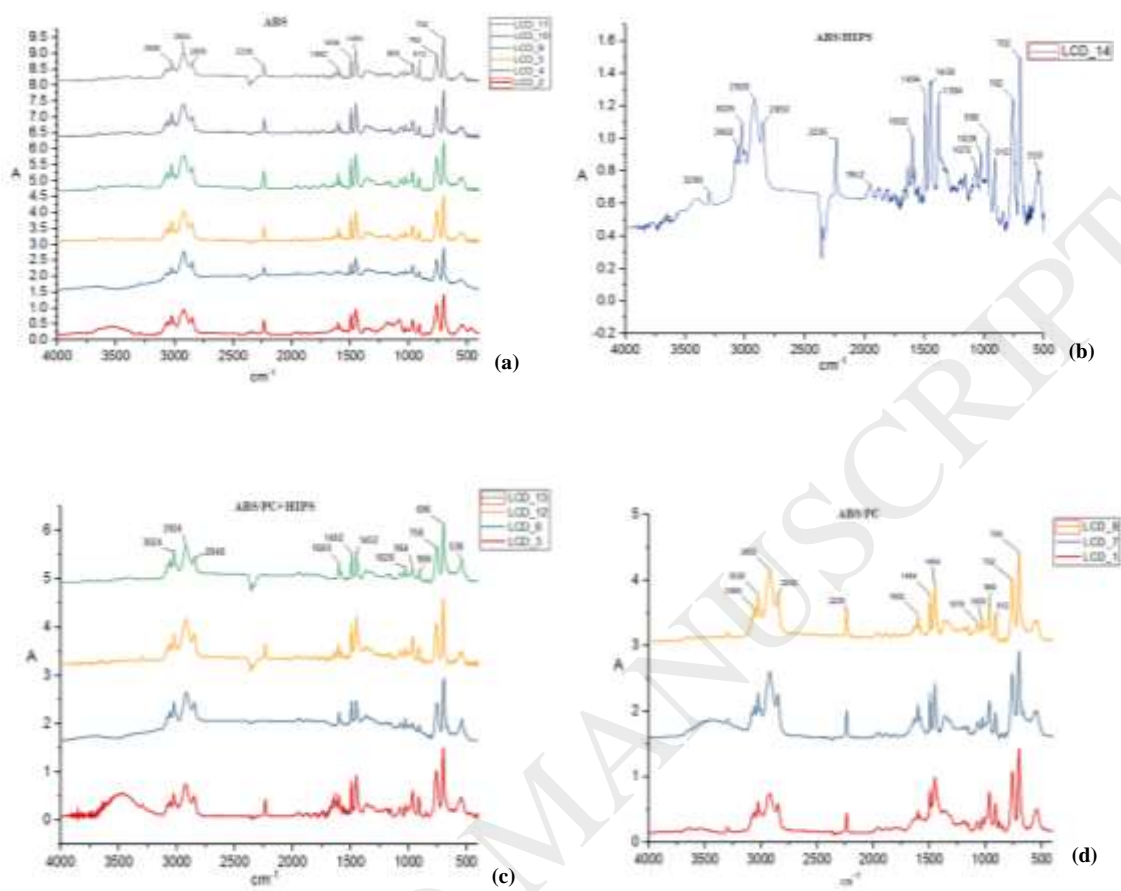
Fig. 1

Fig. 2

Fig. 3

Fig. 4.

Fig. 5.

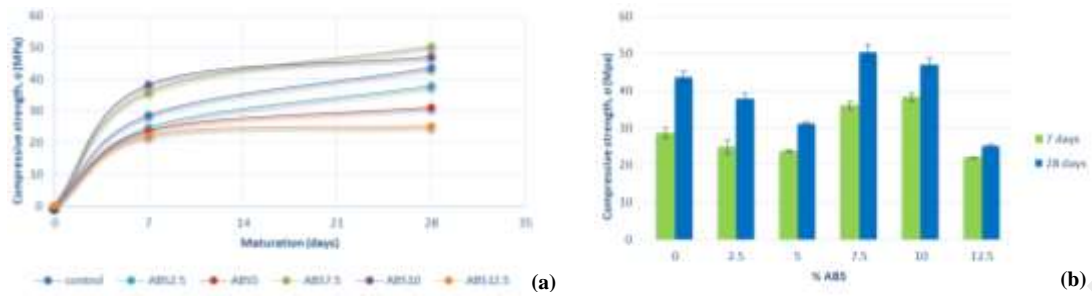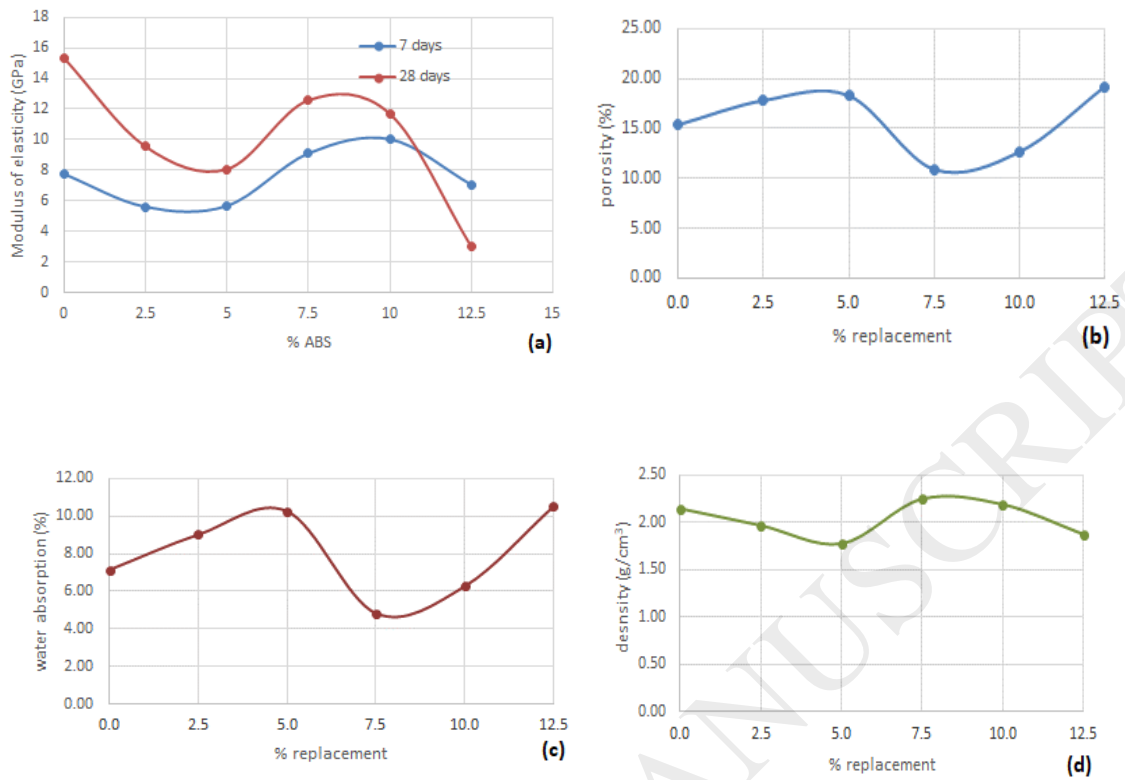**Fig. 6.**

Tables

Table 1. List of examined samples (code named) and polymer types embedded in them.

Table 2. Calorific value (CV) and elemental analysis (EA) of e-plastic samples.

Table 1

Sample code name	Polymer type
LCD_1	ABS/PC
LCD_2	ABS
LCD_3	ABS/PC/HIPS
LCD_4	ABS
LCD_5	ABS
LCD_6	ABS/PC/HIPS
LCD_7	ABS/PC
LCD_8	ABS/PC
LCD_9	ABS
LCD_10	ABS
LCD_11	ABS
LCD_12	ABS/PC/HIPS
LCD_13	ABS/PC/HIPS
LCD_14	ABS/HIPS

Table 2

Sample code name	C%	H%	N%	Calorific value (CV) (MJ/kg)	Elemental analysis (EA) per polymer type according to literature

ABS				
LCD_2	86.38575	9.77375	5.723	34.5
LCD_4	86.604	10.35	3.139	39.5
LCD_5	85.715	10.318	3.149	39.3
LCD_9	85.776	9.649	3.892	39.0
LCD_10	85.376	9.746	3.435	38.9
LCD_11	85.61	8.912	3.006	39.6
ABS/PC+ABS				
LCD_1	76.5075	9.098	4.38	39.4
LCD_7	68.027	9.302	0.986	39.0
LCD_8	86.91	10.80	4.42	39.1
ABS/PC+ABS/HIPS				
LCD_3	80.353	9.415	4.672	39.1
LCD_6	92.742	10.01	-	41.1
LCD_12	86.058	10.14	2.087	39.2
LCD_13	92.841	10.027	-	41.2

Polymer type	C%	H%	N%	Source
ABS	90.07	9.02	0.82	
ABS/PC	80.08	7.32	0.44	[10]
Br - ABS	72.3	6.4	4.1	[26]
ABS	73.3	6	4.5	
HIPS	78.8	6.6	1.2	[27]
ABS	72.3	6.4	4.1	
HIPS	80.5	7.3	0.2	[28]
ABS	70.8	6.7	4.4	[29]
Br - ABS	71.9	6.6	3.9	[30]
ABS	85.9	8	5.08	[31]