INFLUENCE OF ARTIFICIAL WASTE MODIFICATION ON STRENGTH OF CEMENTITIOUS COMPOSITE

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Abstract. *High development of civilisation leads to incremental growth of produced municipal wastes. Despite of introducing restrictions related to municipal selective waste management still great amount of wastes is being landfilled. In countries like India or Philippines engineers started to utilize plastic wastes in extraordinary way using it as an alternative aggregate in cementitious composites. Many researchers tried to improve properties of concrete containing artificial aggregate. This paper analyses various treatment of PET flakes before adding to the mortar mix. Main aim was to modify the surface of PET flakes in order to obtain improved adhesion between artificial aggregate and cement matrix. Mortar mixes contained boiled, stored in alkali and acid solutions and regular plastic particles. This paper presents the results of flexural and compressive strength after 28 days and SEM analysis. Replacing natural aggregate in mortars with 5% PET flakes resulted in decreasing the compressive strength by 7%. Although the surface of PET flakes modified by alkali and acid solutions appeared to become rougher there was no improvement in compressive strength, moreover decreased in comparison to control sample.*

Keywords

Mortar, PET flake, plastic waste aggregate, surface modification.

1. Introduction

High rate of industrial development leads to incremental growth of consumed goods, hence number of municipal wastes significantly increases every year. According to the data provided by Statistics Poland (GUS) total amount of municipal wastes collected in Poland in 2019 was 12.8 million tons. Figure 1 presents municipal wastes collected in Poland per capita [1].

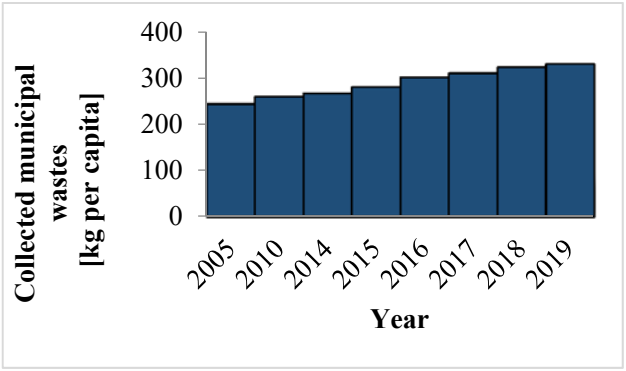

Fig. 1: Amount of collected municipal wastes in Poland per capita [1]

Separate collection amounts to 29% of all municipal wastes. Poland belongs to the group of European countries with ban of landfilling high calorific wastes (over 6000 kJ/kg dry mass), however still over 43% of all the municipal wastes in Poland were disposed to landfills in 2019 [1].

Three popular methods of plastic waste disposal are landfilling, incineration and recycling. Landfilling and incineration can be the source of secondary environmental pollutants. Recycling is more eco-friendly method of waste disposal, although effective recycling of PET requires complex processes of removing the contaminants. Otherwise recycled mate-rial can be less valuable or even be harmful to health. Therefore process of recycling is considered as less cost-effective method in comparison to landfilling and incineration [2], [3].

Figure 2 represents the plastic post-consumer waste rate management for selected European countries. According to the data provided by Conversio Martket & Strategy GmbH [4], Poland in 2018 recycled 27% of collected plastic waste and over 42% was landfilled. In case of waste disposal efficiency Poland is in the 21 position comparing to 30 other European countries.

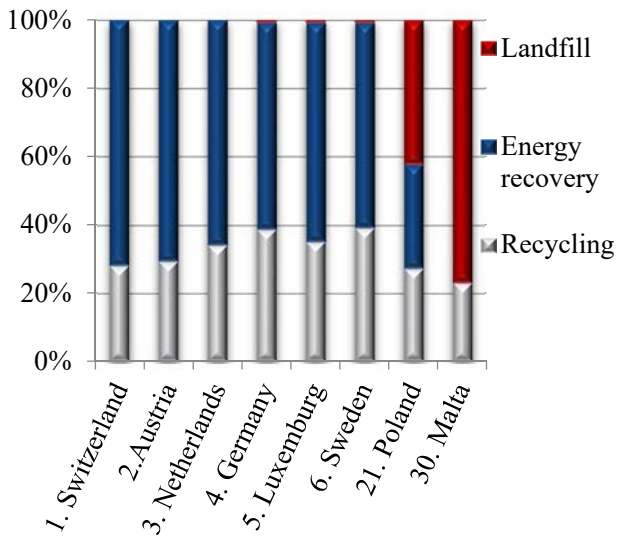

Fig. 2: Plastic post-consumer waste rate of treatment [4]

Effective plastic waste disposal is challenging task for present generation. Currently phenomenon called "micro plastic soup" occurred in the Indian Ocean. The concentration of different size particles was 34000 pieces per square kilometer [5].

For many countries like India, Sri Lanka or Jordan plastic waste constitute a serious threat to the environment [6], [7]. In order to reduce the amount of plastic waste disposed to landfills or abandoned on the streets it is used as a replacement of aggregate in concrete production. Company in Philippines decided to shred plastic bags and used it to produce "eco-bricks", which were the construction material for building a canteen, pavement and clock tower in one school [8].

Substitution of natural aggregate with plastic post-consumer waste is challenging. The main concern of such type of artificial aggregate is that it decreases the mechanical properties of cementitious composites like compressive or flexural strength [9]. The reduction of strength may be caused by poor bond between cement matrix and plastic particles [10]. Researchers tried to modify the surface of PET particles. Scientists used mechanical and chemical methods or introduced extraordinary solutions.

One of the extraordinary method is irradiating plastic by gamma ray. In that research PET flakes absorbed two different amounts of radiation low and high dose. Low dose particles were in the irradiator for 2.9 h and high dose for 28.7 h. Ordinary Portland Cement was the binder, fly ash and silica fume were added in different mixes. For OPC pastes with long and short irradiated PET particles compressive strength increased by about 25% in comparison to non-treated particles. For pastes with silica fume this feature was similar for non-treated (50.4 MPa) and both treated (49.6 MPa and 53.3 MPa). Similar results were for samples with silica fume, particles irradiated with high dose of gamma ray increased strength by 10% in comparison to regular PET. However still there was significant reduction of strength comparing to control cement pastes. For control samples with OPC and mixes with fly ash and silica fume strength was reduced by 23%, 23% and 14% respectively [11].

Modification of plastic surface was also provided by immersing particles in different chemicals. In that paper plastic aggregate was exposed to solutions like water, bleach and alkaline bleach. Compressive strength was higher for samples with PET soaked in alkaline bleach comparing to different treatments. Nevertheless this approach also resulted in strength decrease in comparison to control concrete [12]. PET particles are sensitive to alkaline environment. According to the researches particles degrade in cementitious composites. Alkali solution influenced surface of plastic fibers [13]. Rougher surface of PET particles should improve adhesion between cement paste and plastic aggregate. This research present how immersing PET flakes in acid and alkali solutions influenced strength and structure of cement mortars.

2. Materials and methods

Natural aggregate consisted of standard quartz sand (EN 196-1. As a binder cement CEM I 42.5 R was used. PET particles were obtained from plastic bottles. Recycling company delivered washed and shredded material in a shape of flakes, density of flakes was 1.38 g/cm^3. Standard sand was used as a natural aggregate. Table presents characteristics of cement.

Tab. 1: Cement characteristics

Parameter	***Unit***	***Specification***
Composition	%	95-100 Portland clinker 0-5 other
Compressive strength after 2 days	MPa	≥20
Compressive strength after 28 days	MPa	42.5 ≤strength≤62.5
Chloride content	%	≤0.10

Artificial aggregate was immersed in 3 different solutions namely boiling water, 5% NaOH and 5% HCl. The flakes remained in alkali and acid solutions for 2 and 24 hour and after washing and drying plastic waste substituted natural aggregate in mortar. In the research PET flakes passing sieve 4 mm were substituting natural sand. Flakes were 0.5 mm thick. PET flakes were examined under Scanning Electron Microscope. In order to avoid electron charging part of each flake was covered in silver.

In the research samples were prepared according to the PN-EN 1015-11:2001/A1:2007. Mortar beams were 160x40x40 mm. Control sample standard mortar with water/cement ratio equal 0.5 was the control sample. In this research 5% of volume of sand was replaced by the same amount of PET flakes. Mix design of samples are presented in table 2.

Tab. 2: Mix design of mortars, weight ratio

Type of mortar	Cement	Water	Natural aggregate	PET flakes
Control	1	0.5	3	0
PET			2.85	0.078
PET acid 2h				
PET alkali 2h				
PET acid 24h				
PET alkali 24h				
PET boiled				

Samples were demolded after 24 h and weighted with accuracy 0.05 g. For 27 days samples were cured in water in 20°C. For each mortar mix five samples were prepared, two for SEM analysis and 3 for mechanical strength test.

In order to conduct SEM analysis of mortars samples were cut into smaller pieces and immersed in resin. After hardening samples were grinded and polished for 12 hour in total. Before SEM examination samples were immersed in acetone and treated with sonication device in order to remove impurities. Afterwards samples were examined under SEM JSM-IT100 equipped with aQUAN-TAXEDX (Energy-dispersive X-ray spectroscopy) analysis system. SEM images analysed in the research were taken in Backscattered Electron Composition (BEC) mode at high vacuum, accelerating voltage was 6.0 kV and probe current 50 mA. Magnitude ranged from 50 to 2000.

3. Results

3.1. Density

Fig. 3: shows density of mortars after demoulding; each sample was weighted and measured. The loss of density after adding non-treated PET flakes was 2%. Artificial aggregate immersed in acid for one day and in alkali solution for 2 hour did not affect density.

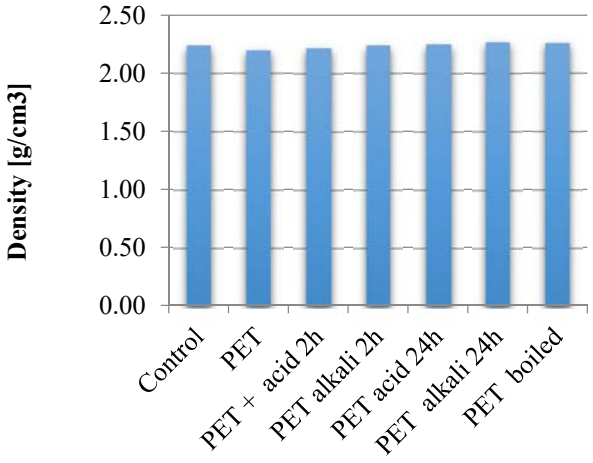

Fig. 3: Density after 24h

Mortars containing flakes immersed in NaOH for one day and in boiling water increased density by 3% in comparison to sample with regular PET flakes. . The results show that addition of artificial plastic waste as a 5% substitution of natural aggregate did not influence the density significantly.

3.2. Flexural strength (PN-EN 1015-11:2001/A1:2007)

Fig. 4: represents flexural strength of mortars after 28 days of curing. Feature decreased by 10% after substitution of natural aggregate with plastic waste aggregate. For mortar containing flakes treated for 2 hour in HCl the loss of strength was 6%. Sample with PET immersed in NaOH for one day obtained result lower by 39% in comparison to standard mortar. Other treating methods did not influence the results significantly comparing to non-treated flakes.

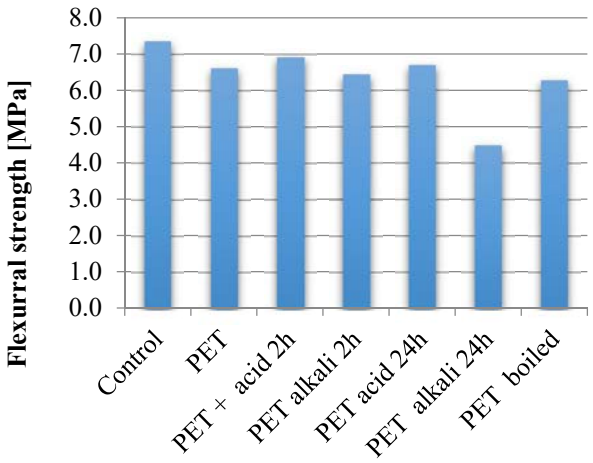

Fig. 4: Flexural strength of mortars after 28 days of curing

3.3. Compressive strength (PN-EN 1015-11:2001/A1:2007)

After performing flexural strength test compressive strength test was conducted. Fig. 5: presents the compressive strength of mortars after 28 days of curing. Substitution of 5% volume of sand with artificial wastes decreased the strength by 7%. Although the surface of plastic flakes was rougher after treating with different solutions it did not result in improvement of compressive strength.

After immersing PET flakes in boiling water and alkali solution (2 hour) the strength of mortar decreased by about 8% in comparison to non-treated particles.

The bar graph in Fig. 5: shows that treatment PET flakes with aggressive solutions, excluding particles immersed in HCl for 24 hour decreased the compressive strength of mortars in comparison to mortar with non-treated PET particles.

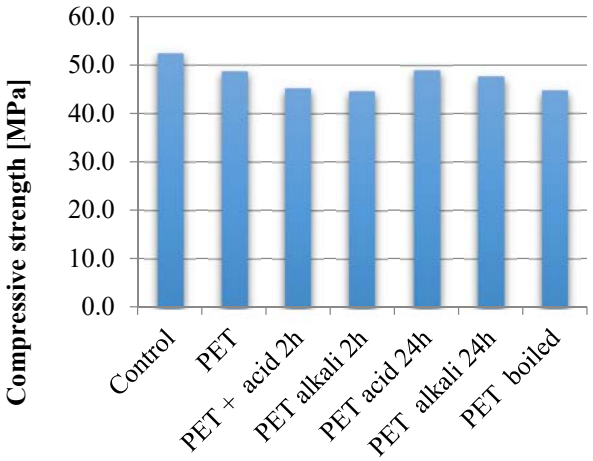

Fig. 5: Compressive strength of mortars after 28 days of curing

3.4. SEM analysis

Figure 6 shows flat surface with scratches after the mechanical process of shredding PET bottles.

After immersing flakes in acid solution there was no additional matter, but the surface become rough and more scratches became visible.

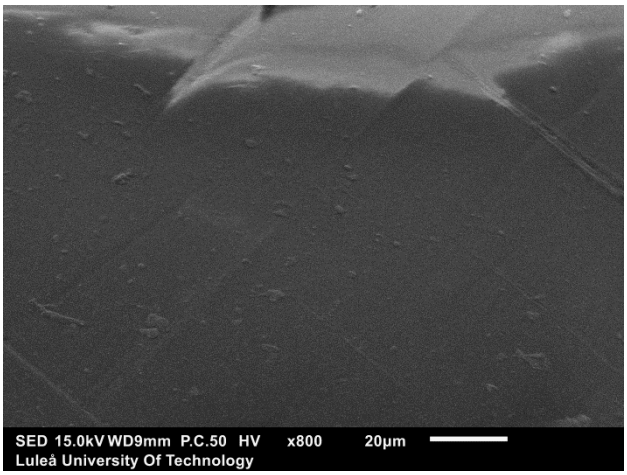

Fig. 6: Non-treated PET flake surface under Scanning Electron Microscope

Figure 7 presents PET flake after immersing in sodium hydroxide for 24 hour. Visually the surface changed in comparison to non-treated plastic flake, therefore EDS analysis was applied.

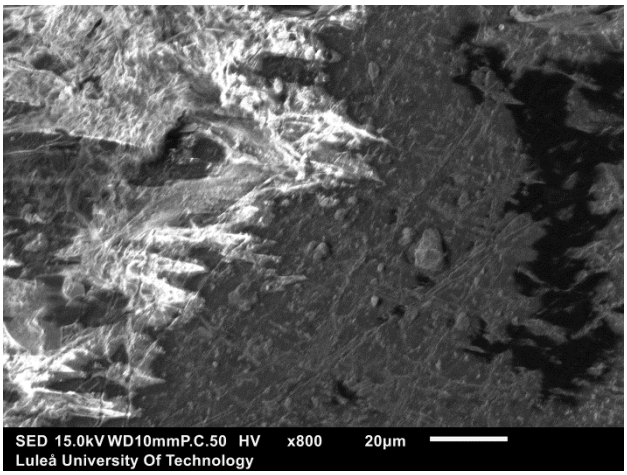

Fig. 7: PET after immersion in NaOH solution for 24 h

Table 3 presents atomic concentration of areas shown in figure 8. Lighter shades visible in the figure contain small amount of silicone. Although flakes were washed and kept in plastic bags still contamination remained on the surface. When magnification decreased the particles like Aluminum, Titanium or Vanadium also occurred on the PET surface as it is presented in table 4.

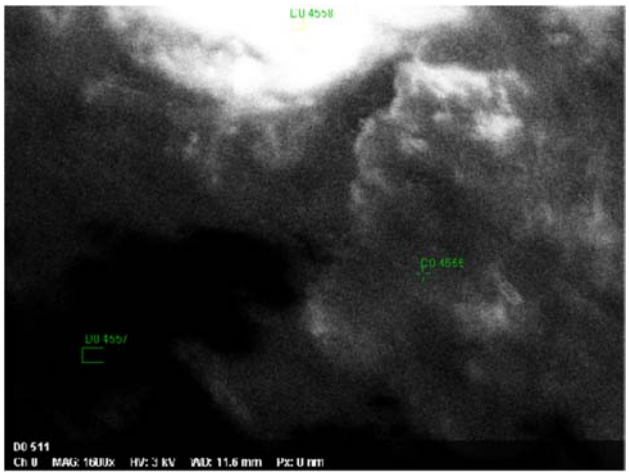

Fig. 8: EDS analysis for non-treated PET, mag. 1600x

Tab. 3: Atomic concentration for non-treated PET presented in figure 8

Spectrum	Carbon	Oxygen	Silicon
D0 4555	78.41	21.03	0.57
D0 4557	74.00	26.00	
D0 4558	78.91	20.54	0.54
Mean	**77.11**	**22.53**	**0.55**
Sigma	**2.71**	**3.02**	**0.02**
SigmaMean	**1.56**	**1.75**	**0.01**

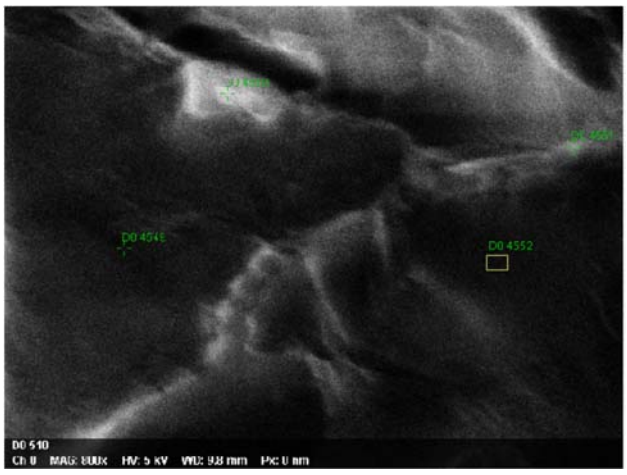

Fig. 9: EDS analysis for non-treated PET, mag. 800x

Tab. 4: Atomic concentration for non-treated PET presented in figure 9

Spectrum	Carbon	Oxygen	Aluminium	Titanium	Vanadium	Rhodium
D0 4549	88.37	11.63				
D0 4550	68.87	19.39			3.00	8.74
D0 4551	70.99	20.60		1.42		6.99
D0 4552	82.14	15.86	0.53	1.47		
Mean	**77.59**	**16.87**	**0.53**	**1.45**	**3.00**	**7.86**
Sigma	**9.25**	**4.03**	**0.00**	**0.03**	**0.00**	**1.24**
SigmaMean	**4.62**	**2.02**	**0.00**	**0.02**	**0.00**	**0.62**

EDS analysis for PET immersed in sodium hydroxide is presented in figure 10. Although there are visible changes in PET surface EDS analysis disproved that PET reacted with NaOH or that the crystals of sodium hydroxide were formed on the surface of plastic flake.

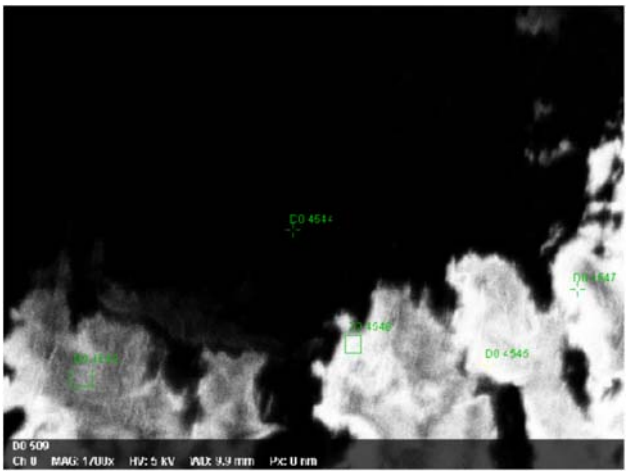

Fig. 10: EDS analysis for PET treated with NaOH, mag. 1700x

Tab. 5: Atomic concentration for areas presented in figure 9

Spectrum	Carbon	Oxygen	Silicon	Scandium
D0 4544	83.58	16.42		
D0 4545	78.92	18.94	0.34	1.81
D0 4546	81.78	18.22		
D0 4547	82.54	17.17	0.30	
D0 4548	80.00	20.00		
Mean	**81.36**	**18.15**	**0.32**	**1.81**
Sigma	**1.89**	**1.41**	**0.03**	**0.00**
SigmaMean	**0.85**	**0.63**	**0.01**	**0.00**

Fig. 11: presents matrix of mortar with addition of non-treated plastic aggregate.

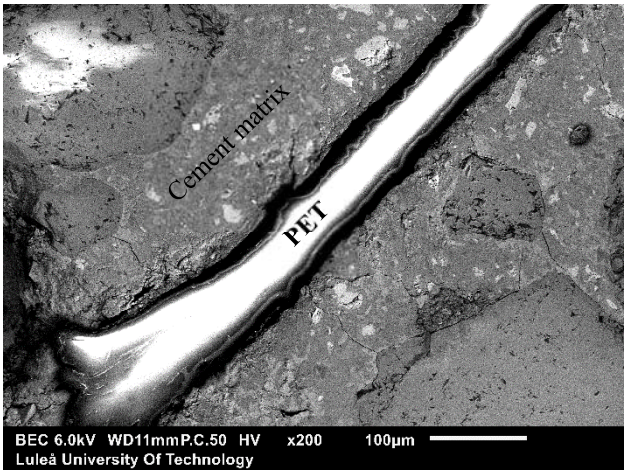

Fig. 11: Mortar containing non-treated PET flakes

Fig. 12: presents interfacial transition zone between cement matrix and PET particle treated with alkali solution for 24 hour. More extensive cracks are visible, moreover some particles attached to the surface of plastic aggregate were separated from cement matrix. Application of EDS showed that separated particles are mainly CSH phase (D0 4603, D0 4606) as it is presented in Tab. 6:.

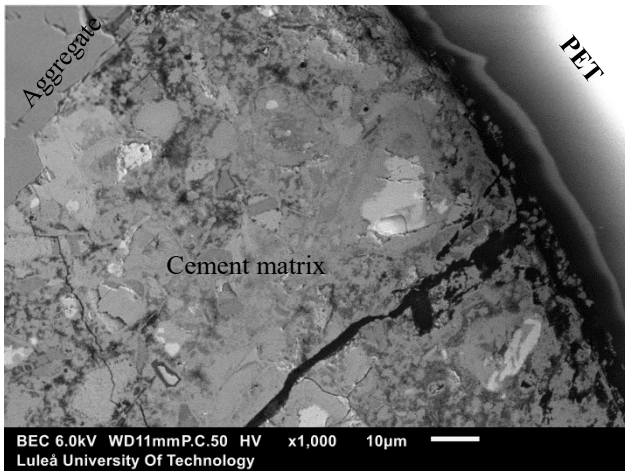

Fig. 12: Mortar containing 5% of PET flakes immersed in NaOH for 24 hour

Fig. 13: shows section between cement matrix and PET flakes immersed in acid solution for 24 hour. Structure is similar to sample containing PET treated with alkali solution, although visible cracks are thinner and appear more frequent.

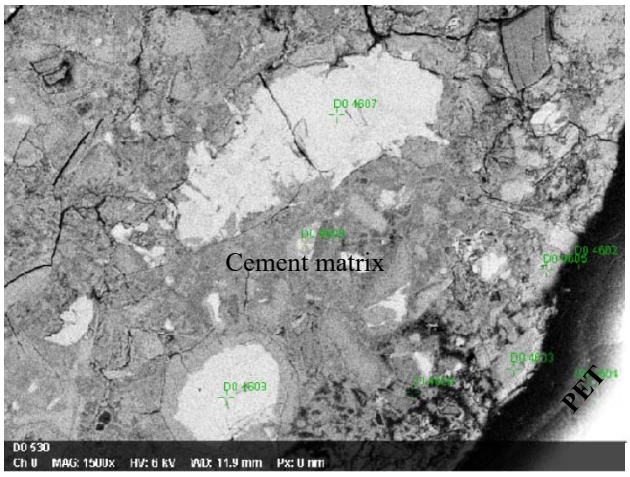

Fig. 13: Mortars containing PET flakes immersed in HCl for 24 hour

Tab. 6: EDS analysis for sample with PET immersed in acid

Atomic concentration [%]

Spectrum	Carbon	Oxygen	Aluminium	Silicon	Calcium	Vanadium	Iron	Tantalum
D0 4602	87.82	9.97		0.24		1.97		
D0 4603	86.48	12.34		0.51	0.66			
D0 4604	86.04	10.29		0.76		2.91		
D0 4605	86.27	12.36		0.60	0.76			
D0 4606	76.47	20.48		1.66	1.39			
D0 4607	72.92	20.87	0.56	1.90	3.68			0.07
D0 4608	75.27	19.50		2.30	2.92			
D0 4609	71.74	24.03		1.86	1.97		0.40	
Mean	**80.38**	**16.23**	**0.56**	**1.23**	**1.90**	**2.44**	**0.40**	**0.07**
Sigma	**6.88**	**5.55**	**0.00**	**0.78**	**1.21**	**0.66**	**0.00**	**0.00**
SigmaMean	**2.43**	**1.96**	**0.00**	**0.28**	**0.43**	**0.23**	**0.00**	**0.00**

SEM images show difference occurring on PET flake surface after immersing it in alkali solution (Fig. 7:). However EDS analysis did not reveal any significant changes in chemical composition of PET flakes. Furthermore no changes occurred in composition of interfacial transition zone in mortars with alkali treated PET in comparison to mortar with non-treated plastic aggregate.

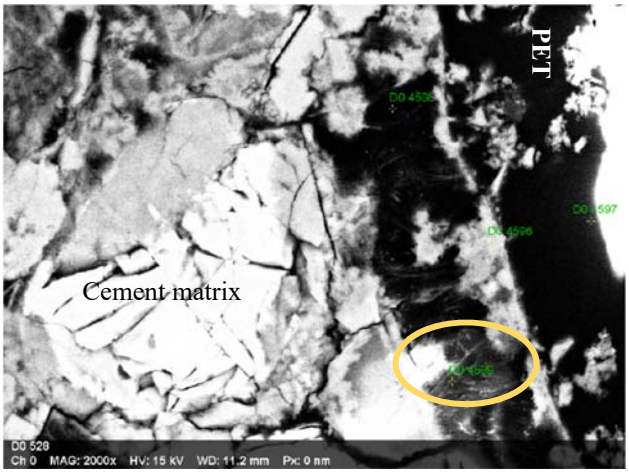

Fig. 14: Mortar containing 5% PET treated with NaOH under EDS analysis

Fig. 14: shows fragment of mortar with 5% of PET immersed in alkali solution examined under EDS. The result of point number 4599 (marked yellow) is presented in table 7.

Tab. 7: EDS analysis results for point number 4599

Element	At. No.	Netto	Mass [%]	Mass Norm. [%]	Atom [%]	abs. error [%] (1 sigma)	rel. error [%] (1 sigma)
Carbon	6	25511	70.41	70.41	78.65	8.83	12.54
Oxygen	8	3474	22.25	22.25	18.66	3.76	16.90
Calcium	20	3633	5.49	5.49	1.84	0.22	4.02
Silicon	14	971	0.72	0.72	0.34	0.07	9.39
Aluminium	13	701	0.52	0.52	0.26	0.06	11.70
Sulfur	16	800	0.61	0.61	0.25	0.06	9.71
		Sum	**100.00**	**100.00**	**100.00**		

4. Conclusion

Although the surface of PET flakes modified with alkali and acid solutions appeared to become more rough there was no improvement in compressive strength. Moreover after immersing plastic aggregate in boiling water, wherein the temperature exceeded glass transition temperature, strength decreased by 8.3% in comparison to non-treated PET mortar. Additionally composition of interfacial transition zone was similar for control mortar and samples containing modified and non-treated PET. This research shows that different approach on PET surface modification needs to be developed in order to classify PET flakes as a valuable replacement of natural aggregate.

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