Polymer modified concrete impact on the durability of infrastructure exposed to chloride environments

Abstract

A limited number of studies are available on the impact of polymer modified concrete on the corrosion effect of Reinforced Concrete (RC) structures, such as bridges exposed to maritime environments or de-icing salts. The aim of this research was to analyse the influence of polymers on the mechanical and durability properties of concrete. Seven types of recyclable thermoplastic polymers PCL, Polymorph, LDPE/EVA granular, LDPE/EVA powder, GPE, N-228 and TPE were used as fine aggregate replacement (3 and 5%). The impact of the molecular weight and structure, size, shape and surface texture of polymers on the fresh and hardened properties of concrete were evaluated. All concretes were designed for a compressive strength higher than 50 MPa. The results show that polymer modified concrete makes the concrete suitable for conventional reinforced concrete manually compacted and heavily reinforced sections with vibrations. Adding granular and powder form of polymers lead to a 2-15% strength increase. For most of the polymers studied, their effect on preventing water migration via capillarity seems to occurr 2-3 days after exposing concrete to water. The substitution of natural fine aggregates with some polymers lead to a reduction of chloride ion migration into the concrete samples, indicating that some of them stop free chlorides inside concrete. 5% LDPE/EVA leads to the higher restricted movement of free chloride migration as the coefficient decreased by 64% in comparison to plain concrete.

Key words: concrete, durability, polymer, maritime, sustainability

1. Introduction

Concrete is susceptible to numerous sources of damage throughout its service life. Cracking-related deterioration of cementitious materials is a serious threat to integrity, durability and safety of concrete-based structures [1]. Reinforcement corrosion in concrete is the main cause of structures-in-service life span decrease and failures, where the carbonation of concrete and the chloride attack are the two most common causes.

Concrete moisture content associated with its porous structure and pore water composition has a direct impact on concrete resistivity, important for the corrosion process [2]. The porous medium (including cracks) created within concrete enables the transport of gases (oxygen and carbon dioxide) water and ions (chloride,), with direct impact on corrosion of structures. The transport is cased by ions concentration gradients, gradients of pressure (gases, water), absolute pressure difference (water, gases), migration and capillary forces. Figue 1 presents the transport process in concrete.

Transport Processes In Concrete

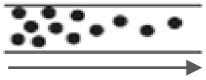
Type	Driving Force	Pores
Diffusion (gases and Ions)	 Concentration gradient dc partial pressure different dp	Filled with air or water
Capillary suction (liquids)	 Surface tension s Contact angle q	Filled with air
Permeation (gases and liquids)	 Absolute pressure difference dp	Filled with air or water

Figure 1 – Transport processes in concrete based on [2].

Chloride uptake is caused by diffuson process and capillarity. The fact that it results in the corrosion process is a different matter. At low relative humidity, capillary suction is more efficient, whereas in wet/humid concrete, diffusion coefficients decrease for gases but play a more important role for the mobility of ions, which is greatest in completely waterfilled pores. The sorption isotherm of the concrete influences the increase and decrease of the diffusion coefficient (Fig.2).

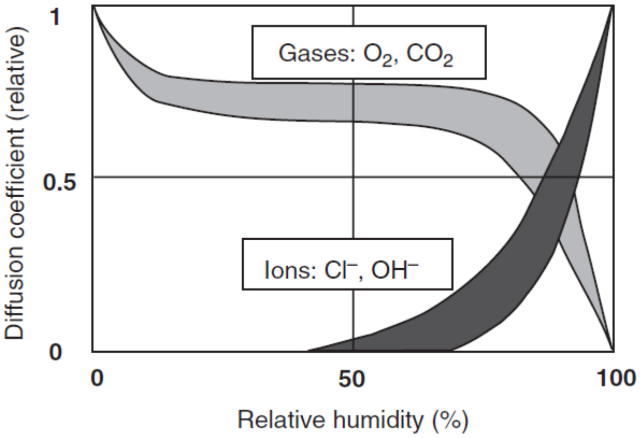


Figure 2 – Diffusion coefficients of ions and gases within the concrete as a function of the relative humidity [2].

Corrosion is a function of the initiation period (where the transport process, chloride ingress and carbonation play important roles towards rebars depassivation) and of the propagation stage, when corrosion starts leading to loss of rebar section. Beside the quality of concrete and depth of cover, the corrosion kinetics depends on the electrochemical reactions, electrolyte resistance influenced by temperature and moisture content (Figure 3).

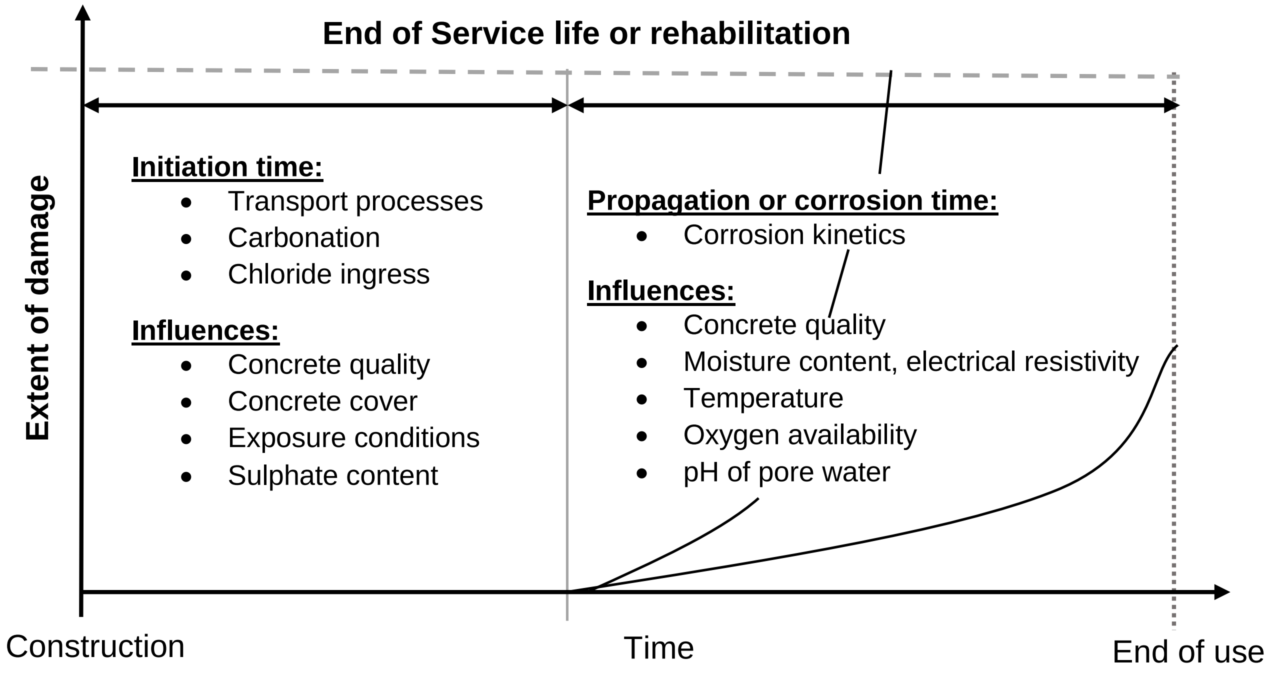


Figure 3 – Initiation and progagation times of corrosion in Reinforced Concrete (RC) [2] [3].

Durability is, therefore, a key important property for materials utilised in the construction and civil engineering industries, emphasising the need for sustainable methods that extend infrastructure service life. Slowing the diffusion and penetration depth of contaminants through the cement matrix is crucial to increase corrosion resistance. Polymer additives might be a solution by filling the voids and/or reacting with the matrix to eliminate or reduce the conduction contact area for corrosive components of the environment. However, there is still a lack of references related to this.

The application of polymers (plastics waste or virgin) in concrete has not yet significantly progressed and received fair publicity in the concrete industry. Therefore, research and innovation to develop new products to replace single-use plastics are necessary to prevent and reduce plastic pollution. The use of polymers as natural aggregate substitution has considerable potential. The most commonly polymers employed are low and high-density polyethylene (LDPE and HDPE), polyethylene terephthalate (PET), polypropylene (PP), polystyrene (PS), and polyvinyl chloride (PVC) etc. The incorporation of polymers can significantly enhance certain properties, as polymer has high heat capacity and low thermal conductivity, high toughness and good abrasion behaviour [5].

Polymers can be thermoplastics or thermosets. Thermoset polymers are those cured by way of heat or chemical reaction, to become a material that is insoluble and infusible. Thermoplastics are composed of molecular chains that are linear and when heated it softens and when it is cooled it is hardened, making them recyclable. There are three basic types of thermoplastic polymers, i.e. amorphous, crystalline and semi-crystalline [6]. Amorphous thermoplastics are normally transparent materials with randomly arranged molecules; examples include poly-methyl methacrylate (PMMA), poly vinyl chloride (PVC), polystyrene (PS), acrylonitrile butadiene styrene (ABS) and polycarbonate (PC). In crystalline thermoplastics the polymers chains present regular arrangement to give translucent materials; this type of polymers has greater resistance to mechanical impact and examples include low-density polyethylene (LDPE), high-density polyethylene (HDPE) and polypropylene (PP). The semi-crystalline type of polymer, present properties that are a combination of those of amorphous polymers and crystalline polymers; this group is represented by polyamide imide (PAI) and polyester polybutylene terephthalate (PBT). There has been extensive research into the use of waste and virgin plastic materials in conventional concrete, but less from a durability perspective of RC. There are usually two forms of polymer that are used in concrete: plastic fibres (PF) used in fibre-reinforced concrete, and plastic aggregate (PA) in the replacement of natural aggregates (coarse or fine).

The following table (Table 1) presents the information of various polymers application from previous studies on plastic waste incorporation in concrete.

Table 1 - Information of various polymers application in concrete composites from previous studies.

Type of plastic used in the concrete	Particle size of plastic (mm)	Replacement%	Properties	References
Metalized plastic waste (fibre)	5 to 20	2.0%, 1.5%, 0.5% and 0.1%.	Oxygen permeability, corrosion, impact load, sulphate attack, weight loss.	[15]
PET (aggregate)	5 to 20	50%, 40%, 30% and 20%.	Workability, compressive strength and density.	[24]
PET (aggregate)	0.15 to 7	15%, 10% and 5%.	UPV and elastic modulus, and flexural and compressive strength.	[45]
Electronic waste plastic (E-waste)	1.86 to 2.78	24%, 20%, 16%, 12%, 8% and 4%.	Compressive strength, permeability and sorptivity.	[46]
PET (aggregate)	0.26 and 1.14 cm (the average size for two fractions)	20% and 10%.	Flexural, tensile and compressive strength, pulse velocity and elastic modulus.	[47]
PVC pipe (aggregate)	≤5 mm	45%, 30%, 15% and 5%.	Dry and fresh density, tensile and compressive strength, water absorption, carbonation, shrinkage and elastic modulus.	[14]
PET (aggregate)	0.5 to 16	15%, 10% and 5%.	Compressive, flexural and tensile splitting strength, .and modulus of elasticity.	[8]

PET (aggregate)	1 to 1.5	5%	Flexural, compressive and tensile strength, pulse velocity and elastic modulus.	[13]

Although using polymer in concrete can be beneficial from the point of view of engineering and the environment, with respect to thermal and mechanical properties, polymers show essential differences from cement and natural aggregates. An experimental research undertaken by Al-Bakri et al [7] claimed that the incorporation of PA in concrete could be used for construction applications as the strength gain is as high as conventional concrete. Nevertheless, the incorporation of PA into the concrete changes the homogeneity and consistency of the fresh mixture properties such as workability. A study conducted by Sakia & de Brito [8] claimed that the shape and surface texture of plastic aggregate impacted the workability of fresh concrete. The smoother surface texture of PA's improved the workability and produced higher concrete slump when compared to control samples. Studies undertaken by Ramesan et al [9] Dhanani and Bhimani [10] and Sakia and de Brito [8] reported an increase in workability while HDPE (high density polyethylene) and LDPE (low density polyethylene) replaced natural aggregates of the concrete at various replacement levels. On the other hand, an opposite outcome of workability was observed by Ashwini et al [11] by detecting a reduction in concrete workability when various percentages of electronic waste plastics (E-plastic) were added into the concrete. The reduction in workability could be due to two reasons first; the angular particle shapes and the sharp edges; second, concrete containing PA has more free water as PA are not able to absorb water [12].

Most of the literature studies claimed that the strength properties (compressive and flexural) of concrete containing PA's are always lower than those of reference concrete, and further reduction with the increase of PA content [11, 13-19]. Mohammed et al [20], studied the effect of PVC plastic aggregate on the properties of concrete at various level of substitution of coarse and fine aggregate. the experimental outcomes showed that there was continuous strength reduction as PVC aggregate inclusion was increased, there was 8% strength loss with 30% aggregate replacement. Albano et al [21] reported that concrete containing 5% and 10% scrap rubber of size 0.59 mm, had a drastic reduction in compressive strength 60% and 88% if compared to conventional concrete sample. Moreover, in terms of flexural strength Farooq et al [19] observed an almost linear reduction of flexural strength with increases in E-plastic waste as partial replacement of fine aggregate (FA) in concrete. The reduction in strength was caused by the weak bonding between the cement paste and the plastic waste surface and the hydrophobic nature of plastic waste that may inhibit the reaction of cement hydration through restriction of the movement of water. Choi et al [22] showed that there were wider gaps in the interfacial transition zone (ITZ) in concrete containing PA's through scanning electron microscopy (SEM) analysis, and this was responsible for the reduced compressive strength of concrete.

Hannawi et al [23] reported an increase in water absorption and apparent porosity with increasing the content of plastic aggregates. Similarly, Islam et al, [24] deducted that the poor bonding between PA and cement paste at the ITZ contributed to increased porosity as well as permeability. However, opposite views were observed by Akcaozoglu et al [25] and Maezouk et al [26], that stated that the concrete permeability and water absorption were enhanced by the addition of PA when compared to the control samples. Redistribution of the calcium-silicate-hydrate (C-S-H) during the hydration of cement enables partial filling of the pores in the ITZ. Aggregates can be interconnected within ITZ, however, this depends on the size and the quantity of aggregate content, which significantly influences concrete permeability and ion transport properties [27].

Furthermore, an increase in resistance to the permeability of the chloride ion with increase of PA content was noted by Kou et al [14]. Similarly, Fraj et al [28] observed a lower chloride diffusion coefficient value. Opposite findings were reported by Silva et al [29] that the chloride permeability of concrete containing waste PET plastic as FA and CA (coarse aggregate) was higher than that of conventional concrete. A greater coefficient of migration was observed with increase of PA content, due to the increase of pore structure in the concrete.

The concrete curing condition also have a great impact on the durability and mechanical properties of concrete containing plastics [29, 30]. Concrete made with plastic aggregate is able to prevent or divert micro-crack propagation and enhance the strength, which is of great practical importance [23, 31].

Further research is needed to elucidate how the properties of polymers, including particle shape, size and surface texture influence the properties of both fresh and hardened concretes. Therefore, this research study aimed to analyse the influence of different types and concentration of polymers on the concrete properties related to durability and service life impact on RC structures.

2. Materials and sample preparation

2.1 Cement

Type one ordinary Portland cement (CEMI) was used in accordance with BE EN 197-1 [32] , with 52.5 N of strength class. The specific gravity was 3.13g/cm^3. The chemical composition of OPC is shown in table 2.

Table 2 - Chemical Analysis of OPC.

Element %	OPC
Fe_2O_3	2.62
CaO	68.91
SiO_2	19.99
MnO	0.06
Al_2O_3	4.32
MgO	1.21
TiO_2	0.18
Na_2O	0.22
K_2O	0.73
L.O.I.	4.00
Acid soluble Cl	0.07

2.2 Fine aggregate

The sand used originated from Tarmac Plant, UK,according to BS EN ISO 9001. The particle size distribution (PSD) for the used sand is shown in the figure 4. The specific gravity of sand was 2.73g/cm^3 with a water content of 0.4%.

2.3 Coarse aggregate

The aggregates used was supplied by Specialist Aggregate and Travis Perkins in the UK, the aggregate sizes were 10 and 20 mm and the particle size distribution (PSD) shown in figure 4. Aggregate 10mm presents a specific gravity of 2.80 g/cm^3 and a water content of 0.1%. Aggregate 20mm presents a specific gravity of 2.50 g/cm^3 and a water content of 2.0%. Determination of Particle Size Distribution was done according to BS EN 933-1.

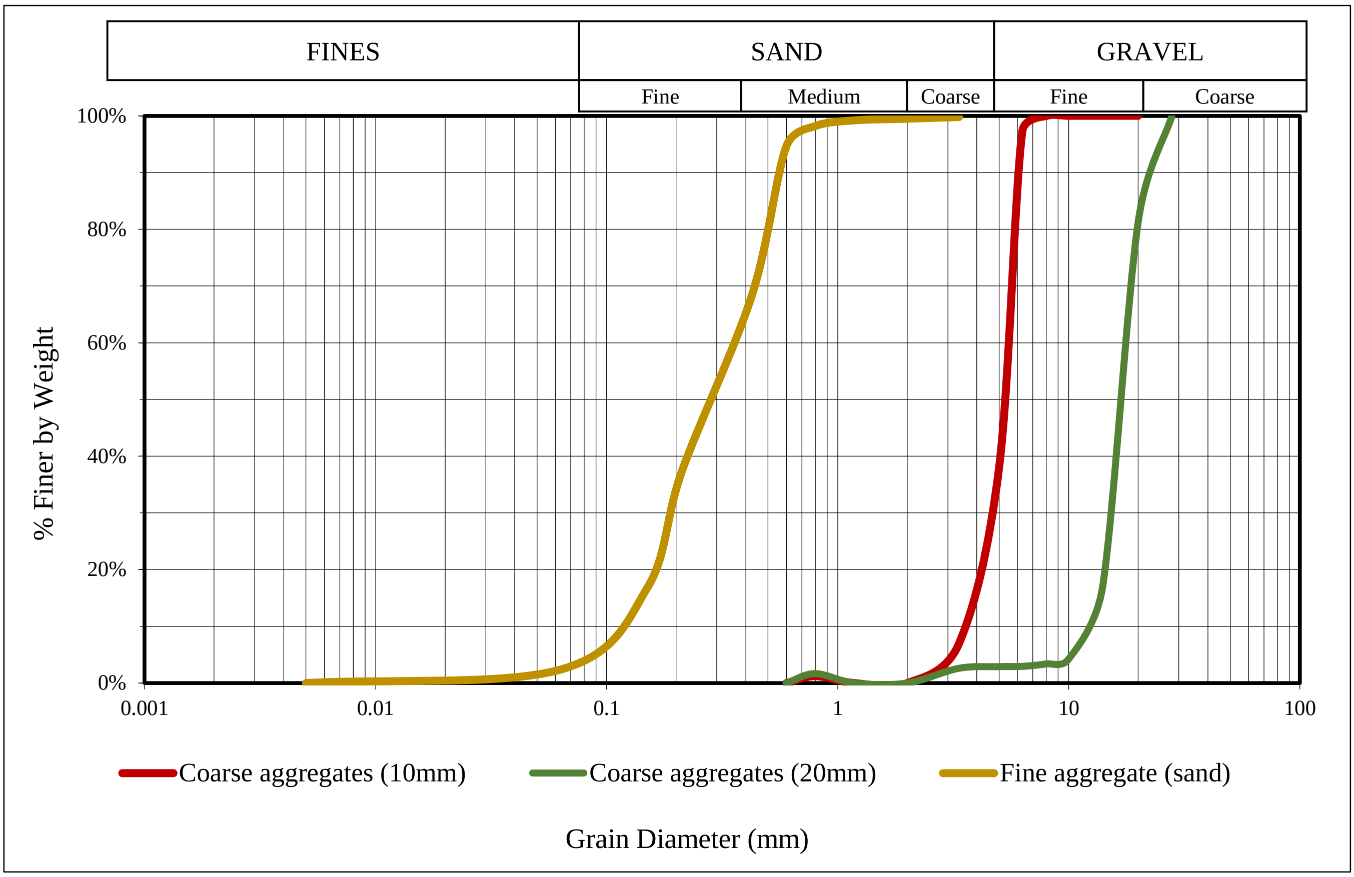
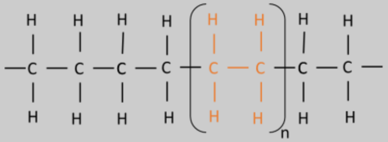
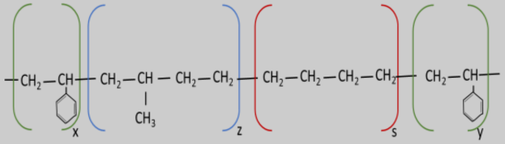
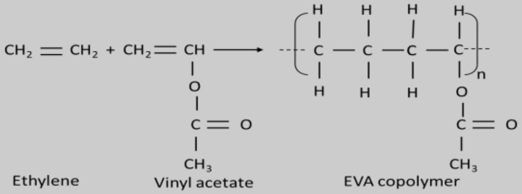


Figure 4 – Particle size distribution for coarse and fine aggregates.

2.4 Polymers

Seven types of thermoplastic polymers were selected for this study, as they are potentially recyclable, abundant in the built environment (e.g. plastic bags, bottles, storage containers, utensils) and have not been considered before in the durability of infrastructures. The polymers are: ε-caprolactone (PCL), Polymorph, low density polyethylene copolymer with Ethylene-vinyl acetate (LDPE/EVA) granular and powder, general polyethylene (PE), plastomer with ethylene-vinyl acetate EVA (N-228) and thermoplastic elastomers (TPE) (SEEPS). The properties of the polymers are shown in the Tables 3, 4 and 5.

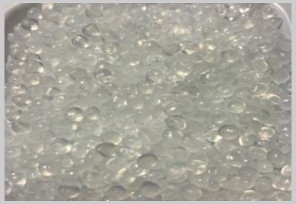
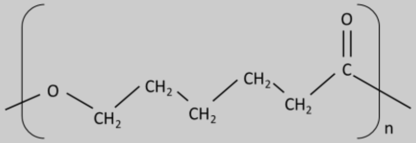
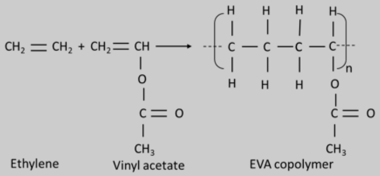
Table 3: Properties of thermoplastic polymers used in the concrete.

Properties	**Polyethylene PE**	**Thermoplastic elastomer TPE (SEEPS)**	**Low density polyethylene/ Ethylene-vinyl acetate LDPE/EVA**
			
Density (g/cm^3)	0.935	0.88	0.90
Specific Gravity	0.926-0.940	0.91	
Type	semi-crystalline.	TPEs are made of polymers that have both hard (semi-crystalline) blocks and soft (amorphous) blocks along the backbone. The soft blocks are left to form amorphous rubbery domains that provide the elastomeric bridges between the crystalline domains	LDPE is semi-crystalline and is VA amorphous
MFI g/10	7.0	<0.1	7.5
Molecular weight (Da)	280,000	90,000	*
Molecular structure			
Monomer	Ethylene (olefin) monomers	Styrene-[ethylene-(ethylene-propylene)]-styrene block copolymer	Co-polymerization of ethylene (olefin) monomers/ethylene and vinyl acetate (VAc).
Monomer Bond type	Double bond	Single bond	Double bond
Resistance to chemicals	Sodium Chloride, Sodium Salts, Sea Water, Potassium Chloride, Iron Salts, Sodium Nitrate, Silver Nitrate,	hydroxides, methanol and ethanol, absorb oil, fats, aliphatic hydrocarbons	Calcium hydroxide, Calcium oxide, Carbon dioxide, Calcium chloride, Ferrous chloride, Potassium chloride, Silver nitrate, Sodium Nitrate,

	Calcium Hydroxide, Sodium Hydroxide, Ferrous Salts.		Potassium Chloride, Sodium Hydroxide, Sodium Chloride
Melting point ˚C	124-134	*	140-220

*(information not provided by the suplier).

Table 4: Properties of thermoplastic polymers used in the concrete

Properties	**ε-Caprolactone PCL and Polymorph**	**LDPE/EVA (VA content 4.5%)**	**Plastomer Ethylene (N- 228)**
			
Density (g/cm^3)	1.07-1.20	0.928	0.902
Specific Gravity	1.15	*	0.902
Type	Semi-crystalline, hydrophobic, biodegradable linear polyester.	semi-crystalline	semi-crystalline
MFI g/10	4.03-2.01*	2.0	10
Molecular weight (Da)	80,000	High molecular weight as the MFI is low	*
Molecular structure			*
Monomer	Linear polyester derived from caprolactone monomer	Co-polymerization of ethylene (olefin) monomers/ethylene and vinyl acetate (VAc).	Plastomers are ethylene alpha olefin copolymers that bridge the gap between elastomers and plastics
Monomer Bond type	Double bond	Double bond	*
Resistance to chemicals	PCL exhibits excellent chemical and solvent resistance.	Calcium hydroxide, Calcium oxide, Carbon dioxide, Carbon dioxide, Calcium chloride,	

	PCL is soluble in almost all aromatic, polar, and chlorinated hydrocarbons and insoluble in aliphatic hydrocarbons, alcohols, and glycols. PCL exhibits mechanical properties similar to those of other conventional nonbiodegradable synthetic polymers. High molecular weight PCL has mechanical properties and oxygen permeability comparable to polyethylene (PE).	Ferrous chloride, Potassium chloride, Silver nitrate, Sodium Nitrate, Potassium Chloride, Sodium Hydroxide, Sodium Chloride	
Melting point ˚C	58-60 melts in water	104	*

*(information not provided by the suplier).

Table 5: Polymers' solubility in water, size and percentage of sand replacement for concrete mix.

Polymers	Type	Water soluble	Size (mm)	Source	Fine aggregate Replacement by mass
PCL	granular	Yes at 62 ˚c	2	Plastiserve	3%
Polymorph	granular	Yes at 62 ˚c	2	Coscraft	3%
LDPE/EVA	granular	No	3	Plastiserve	3%
LDPE/EVA	powder	No	≥ 1.18	Matrix polymers	3% and 5%
GPE	powder	No	≥ 1.18	Matrix polymers	3% and 5%
N-228	powder	No	≥ 1.18	Matrix polymers	3% and 5%
TPE	powder	No	≥ 1.18	Plastiserve	3% and 5%

The following figure presents the grain size distribution of the polymers with non-monogranular dimensions.

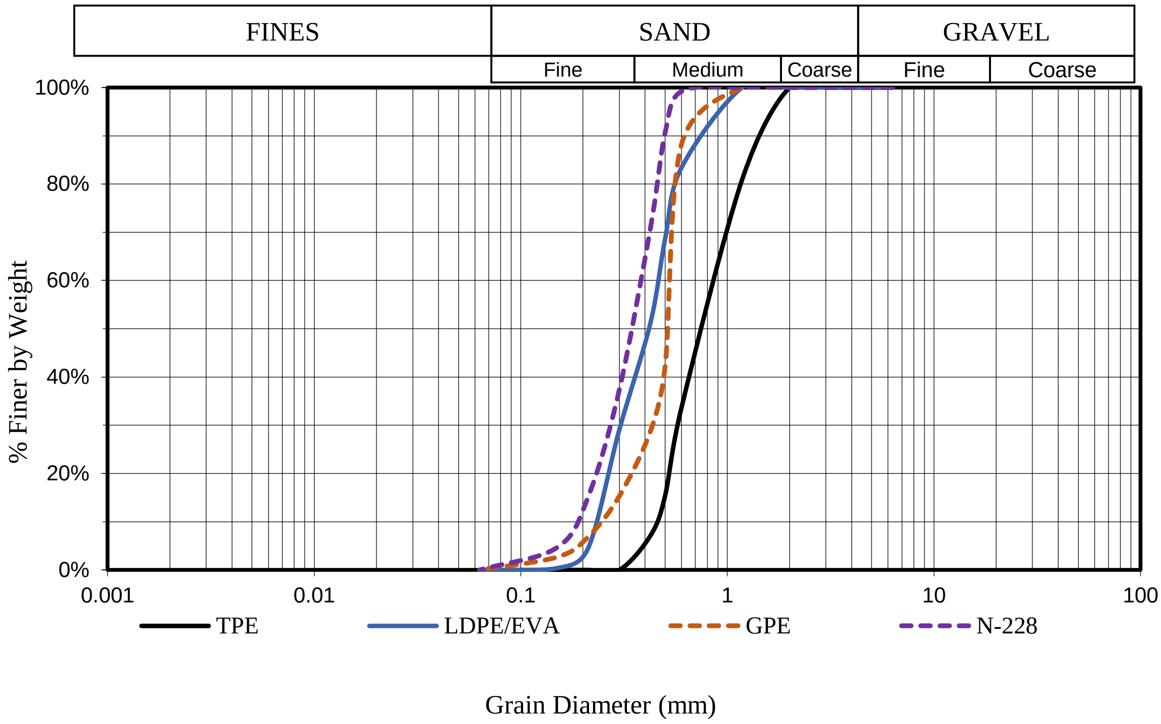


Figure 5 – Particle size distribution of TPE, LDPE/EVA, GPE and N-228.

The experimental plan was designed to assess all concrete samples containing polymer beside the control sample (without polymer). The conducted experimental work and outcomes are detailed below.

2.5 Concrete mix design and sample preparation

The concrete mix were designed and have been cast with Ordinary Portland Cement OPC, along with replacing fine aggregate at the rate of 3% and 5% with the individual thermoplastic polymers. This replacement ratio decision was followed by the literature findings, with a concern on not worsening fresh and hardened state properties of concrete.

In order to avoid migration of floating polymers within the concrete mix, due to their lower specific gravity, compromising concrete homogenisation, compressive strength and increase the scattering of results, Das et al [41] stated that the optimum replacement percentage should be between 2% - 6% as this order would provide higher concrete compressive strength. This is due to the lower density of polymers when compared to natural aggregate. This would mean that the replacement quantity is much more than natural aggregate, occupying bigger volume in concrete. Therefore, with increasing the replacement percentage the strength steadily decreases. Table 6 shows the concrete mix design.

Table 6: Concrete mix design.

Concrete Composition	Kg/m^3	Quantities (kg)
CEMI 52.5N	450	9.54
Aggregate 20mm	712	15.1
Aggregate 10mm	610	12.9
Sand	335	7.1
Water	180	3.8
w/c	0.4	
Polymers	10.05	3% and 5%

2.6 Casting and curing of concrete specimens

Eight series of concrete compositions were mixed and cast in accordance with BS 1881-113: 2011 [33]. For each concrete composition, three cubes 100 x 100 x 100 mm, one beam 100 x 100 x 500 mm and three cylinders with a diameter of 100 mm and a height of 200 mm prepared and casted with water to cement ratio (w/c) 0.40, as shown in Figure 6 below. After 24 hours the samples were demoulded and cured in warm water at 40˚C for a week then they were moved to normal water at temperature ≈ 20˚C.

Figure 6 – Casting of concrete cubes and cylinders.

3. Tests and methods

3.1 Water loss analysis of polymers

The water loss analysis of polymers were undertaken in order to investigate the absorption capacity of different types polymers. Initially 5 grams of each polymer with 20 ml of water were placed in lidded petri dish and were kept at atmospheric temperature for 2 hours (with closed lids). The petridishes were placed in oven at 40 ˚C for 96 hours, then weighing the polymers to analyse how much water evaporated from the polymer.

3.2 Slump test

This research study recorded slump test results, a way to assess the consistency of fresh concrete, following to BS EN 12350-2:2009 [34], for all concrete mixtures during casting. The slump test is sensitive to changes in concrete consistency between a slump value of 10 and 220 mm, enabling to classify concrete using different consistency classes following BS EN 206-1.

3.3 Compressive strength

In compliance with BS EN 12390-3:2009 [35], the structural properties of the 100 mm concrete cubes are subjected to strength testing. In a compression testing machine in compliance with EN 12390-4, samples were loaded to failure.

3.4 Water absorption via capillary

A capillary test was carried out as per EN 1015-18 [36]. Samples were placed in oven at 70°C to dry for a period of 8 days until the mass change was less than 0.1%. The concrete cylinders with 50 mm thickness were placed in 5 mm of water over the absorbent paper within an airtight container to keep hydrothermal conditions consistent as shown in figure 7. Weight of the samples were registered at 0’, 5’, 15’, 30’, 1h, 2h, 3h, 21h up to 28 days, until the water absorption reached the asymptotic value.

Figure 7 – Water absorption concrete via capillary.

3.5 Open porosity

Porosity test carried out in accordance with BS EN 1936:2006 [37]. The volume percentage of open porosity in concrete specimens is calculated using the following equation (eq.1).

$$Open\ Porosity = \frac{M_3 - M_1}{M_3 - M_2} \times 100 \quad (1)$$

Where M_3 is the mass (g) of the sample saturated with water (using a vacuum pump for replacing air in the voids with water), M_1 is the mass (g) of the sample after drying at 70°C for 8 days; and M_2 is the mass (g) of the sample in hydrostatic conditions (saturated and submerged in water). The cylinder samples of dimension 100mm ø and 50mm length shown in Figure 8 were cut into 4 pieces then 3 of them used in each composition.

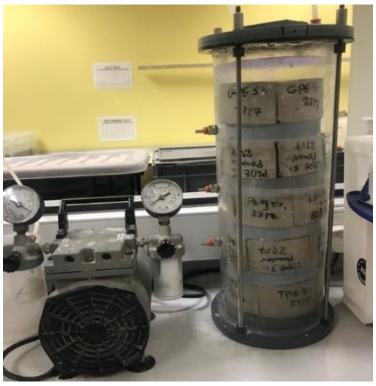


Figure 8 – Concrete open porosity test.

3.6 Chloride migration tests

Chloride migration experiment was conducted as per NT BUILD 492 standard [38], for all concrete samples (with and without polymer) with a thickness of 50 mm and a diameter of 100 mm. The concrete cylinder samples were vacuumed for three hours then flooded with a saturated calcium hydroxide Ca $(OH)_2$ solution as shown in Figure 9(a). The samples remained in the vacuum for an additional 18 ± 2 hours.

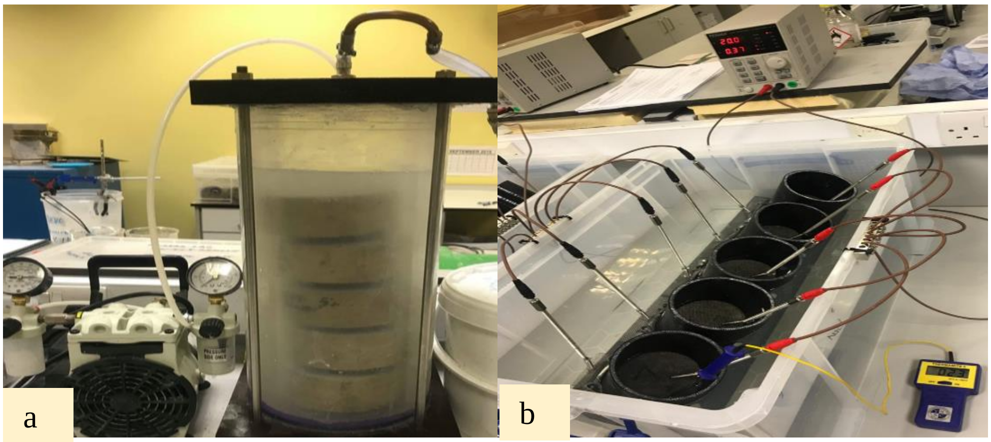


Figure 9 – (a) Preconditioning samples in the vacuum chamber. (b), voltage applied for 24 hours.

After preconditioning, samples were tightly sealed in rubber sleeves exposing only the top and bottom faces. The bottom face exposed to a 10% sodium chloride (NaCl) catholyte solution (by mass) and the top face exposed to a 0.3 sodium hydroxide (NaOH) anolyte solution shown in Figure 9 (b). The power source and initially 30 V was applied, current is measured then adjusted on the final V for 24 hours duration. The concrete cylinders were split axially into two pieces by using hydraulic compression machine, then silver nitrate solution was sprayed on the surface to measure the chloride penetration into the samples.

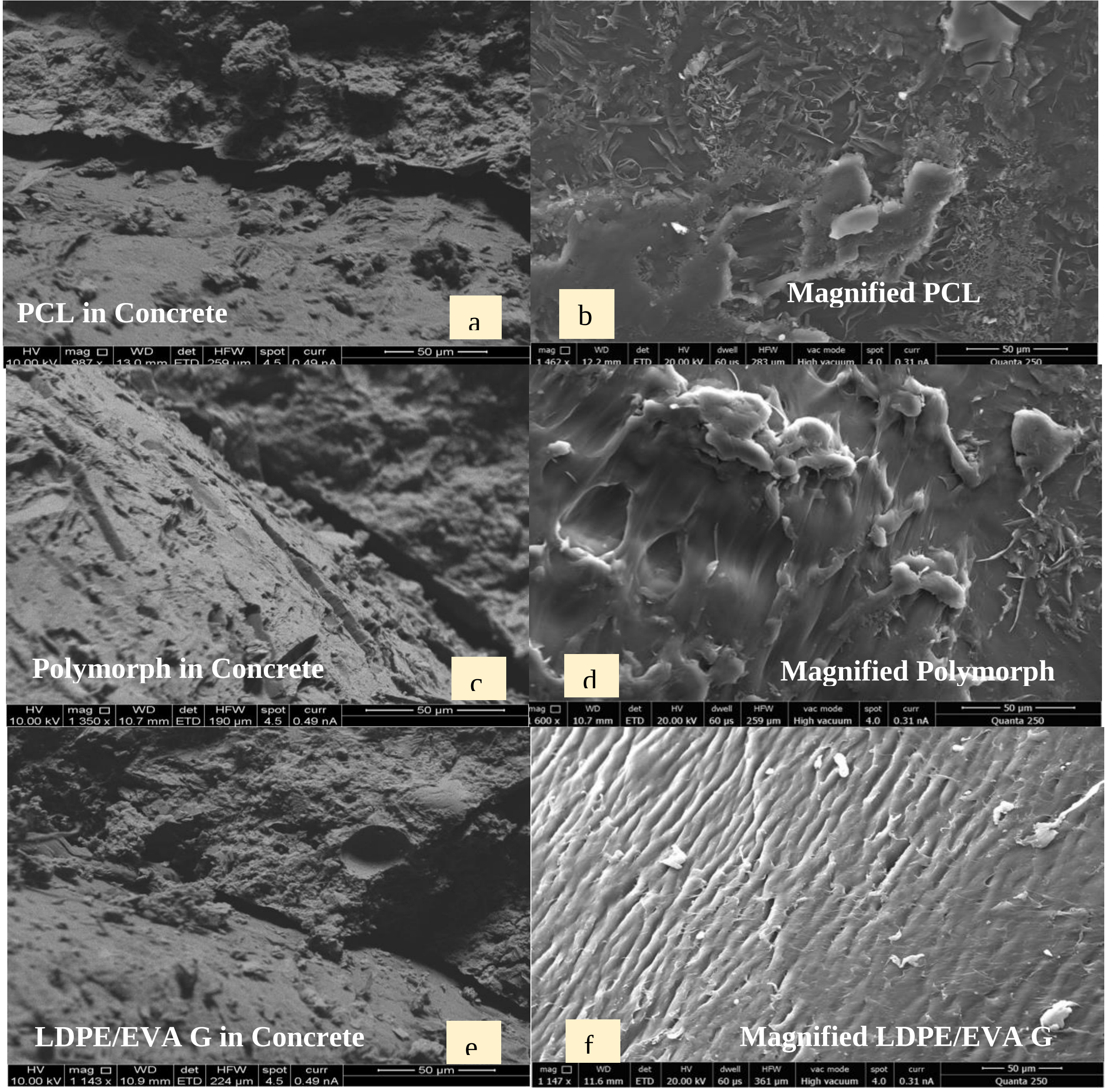
3.7 Scanning electron microscopy (SEM) analysis

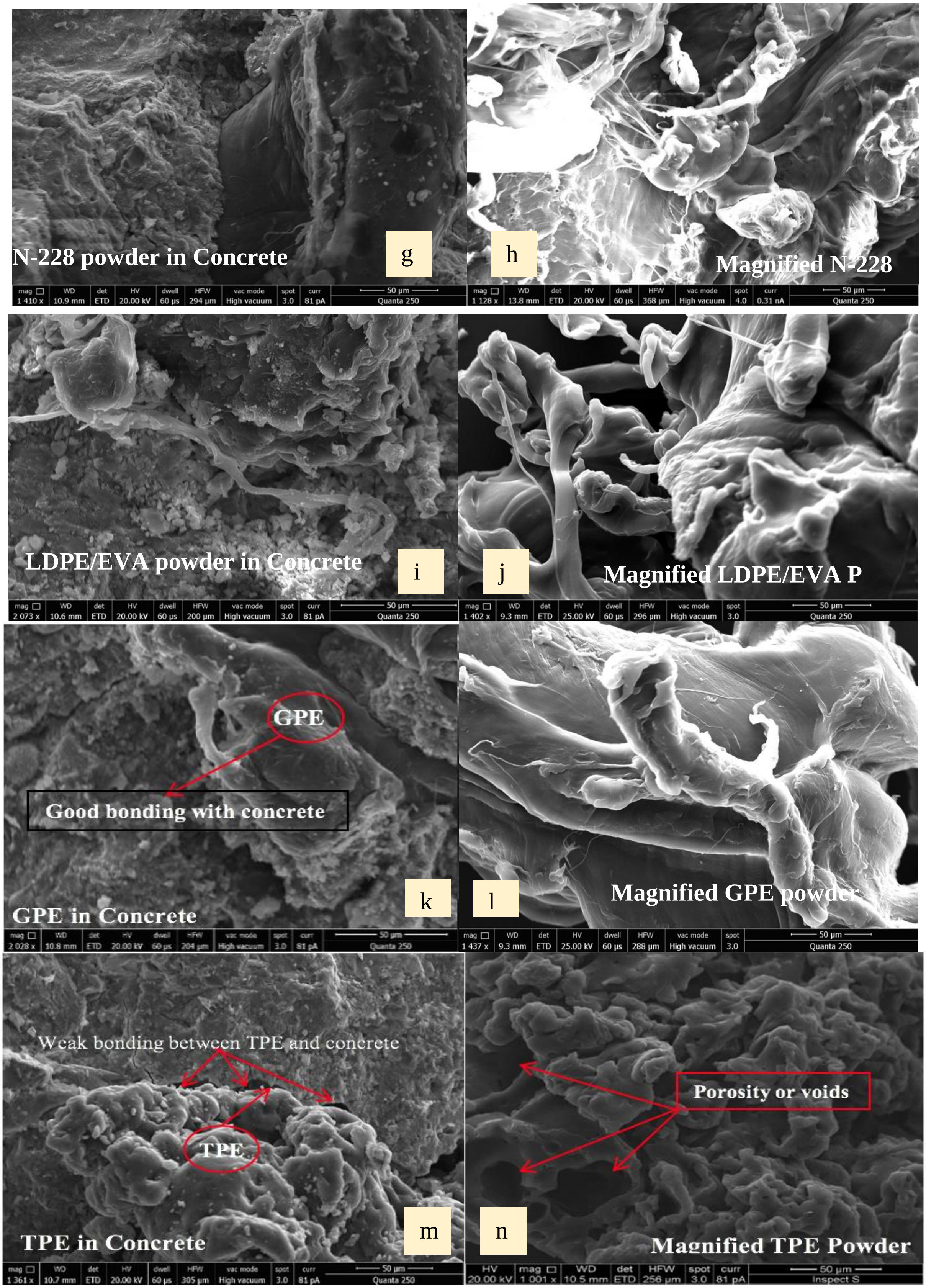
FEI Inspect S SEM variable vacuum. Kv range 0.1-30kv. With back-scattered detector as well as secondary electron detector and x-ray detector was used to analyse the microstructure of polymers and concrete containing polymer with a focus on the matrix of polymer adhesion. Each concrete sample was crushed and coated in gold in order to investigate the bonding characteristic between the cement paste and the polymers.

4 Results and discussion

4.1 Scanning electron microscopy (SEM) analysis

The following Figures 10a-10o present the SEM analysis for polymers in hardened concrete (a, c, e, g, i, k, m), polymer alone (b,d,f,h,j,l,n) and plain concrete (o). Micro-cracks were observed in ordinary concrete without polymer additions (Fig 10o). However when polymers are mixed, the SEM test results shows that there is a good bounding between the tested polymers and the concrete matrix, preventing the formation of micro-cracks in concrete. It was also observed that the surface texture of polymer influences the bond between cement paste and polymer. The smoother surface of LDPE/EVA G (granular) leads to poor bonding compared to rough surface texture (as the one presented by GPE).




N-228 powder in Concrete
g
h
Magnified N-228
LDPE/EVA powder in Concrete
i
j
Magnified LDPE/EVA P
GPE
Good bonding with concrete
GPE in Concrete
k
l
Magnified GPE powder
Weak bonding between TPE and concrete
TPE
TPE in Concrete
m
n
Porosity or voids
Magnified TPE Powder
50 µm

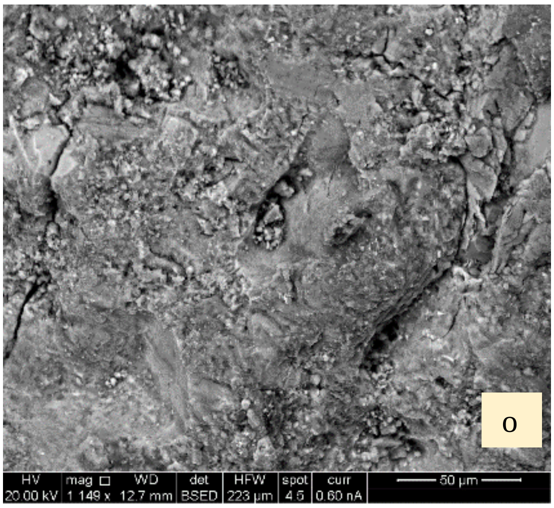


Figure 10: SEM analysis for polymers in concrete (a, c, e, g, i, k, m),polymer alone (b,d,f,h,j,l,n) and plain concrete (o).

4.2 Water loss analysis of polymers

The results presented in figure 11 show that the majority of polymers when exposed for 96h at 40 °C, loose absorbed water. However, for TPE the water seems to escape slower, enphasing the existence of higher chemical bonding with water. From figure 10(m&n) it can be seen that the TPE show a rougher surface, which increases the area of polymer in direct contact with water. The molecular weight tend to affect the surface tension of polymers. For polymers with high molecular weight this effect can be neglected, but not for polymers with low molecular weight. In comparison to other polymers, TPE present a low molecular weight, which associated with a low diffusion rate might explain this behaviour.

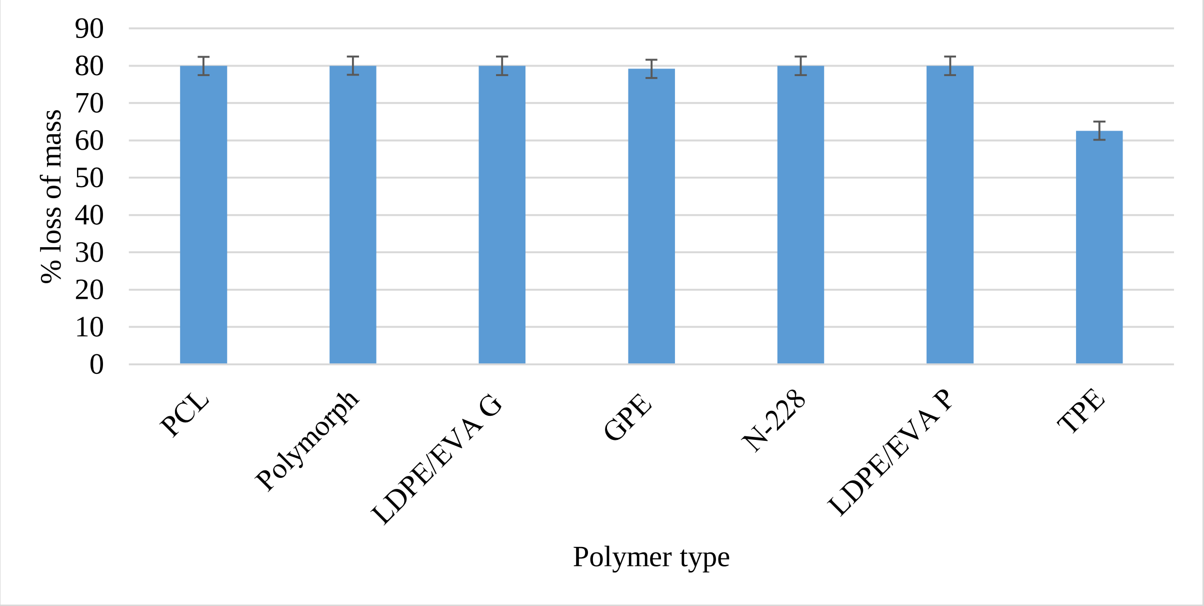


Figure 11 – Water loss analysis for polymers after 96 hours at 40 °C.

4.3 Workability of concrete containing polymers

The workability of concrete can have a significant impact on the strength, consistency as well as the durability of the hardened material, and it is directly affected by the w/c, PSD and shape of the particles. Figure 12 shows the experimental resuts of the slump test. The concrete mixtures have high workability with a consistence class of S3, making the concrete suitable for normal reinforced concrete manually compacted and heavily reinforced sections with vibrations. In general, all the polymer compositions increase the slump by 3% in comparison to control mix. The slightly improved workability of concrete can be explained on the basis of the particle size, shape and smoother surface texture of polymers particles compared to the natural fine aggregate. The polymer size has a great influence on the concrete workability; the mixtures with LDPE/EVA with larger particle sizes of 3 mm showed lower slump when compared to the mixtures containing finer polymers sizes of 2 and 1.18 mm. Furthermore, through SEM analysis it was observed that the surface texture of polymer influences the bond between cement paste and polymer (see figure 10). The smoother surface have poor bonding compared to rough surface texture. In this S3 slump class, 3 different groups were observed: Group 1 (140mm): Control and 3%LDPE/EVA G concrete compositions; Group 2 (145mm): 5%GPE, 3%TPE, 5% TPE, 3%LDPE/EVA P, 5%LDPE/EVA P and 5%N-228 compositions; Group 3 (150mm): 3%PCL, 3%GPE and 3%N-228.

Concrete workability plays a vital role in development of microstructure and consequent development of hardened concrete properties. Some amounts of free water were observed after casting concrete with polymer. Figure 11 intends to show the water loss/ absorption by each type of polymer in order to understand their influence on concrete workability. The findings of the current study showed a clear potential of substituting fine aggregate with polymers in improving fresh concrete workability. Thus, the findings of this research are in agreement with the findings of Ramesan et al [39] and Dhanani and Bhimani [10]. Sakia and de Brito [8] stated that the shape and surface texture of polymers have direct influence on the workability of fresh concrete. The smoother surface texture can improve the workability and produce higher concrete slump when compared to control samples.

However, 3% LDPE/EVA granular presented the same slump value as control this could be due to the particle size, which was bigger than other polymers as they did not melt during the concrete mixing. Albano et al [40], claimed that the bigger particles tend to decrease concrete workability. The primary reasons for the lower slump were the angular shape of the polymers and its sharp edges. Also, from the above discussion, it can be seen that the size greatly affects concrete's workability.

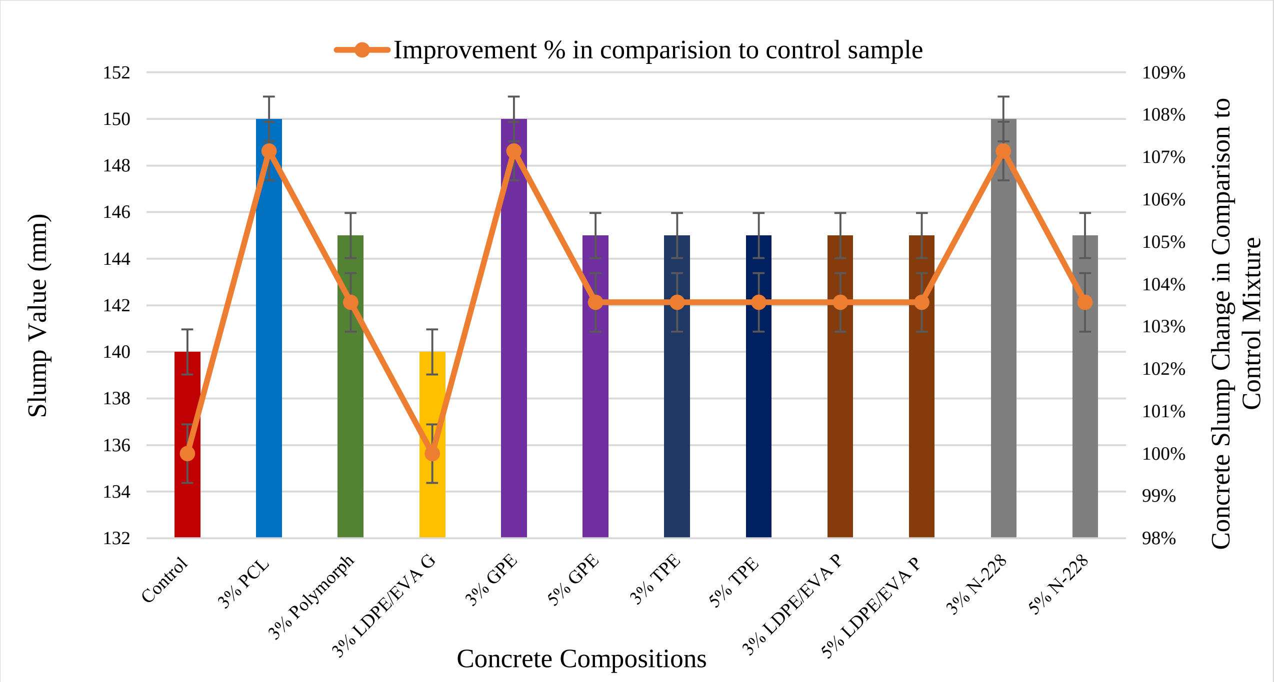


Figure 12 – Slump values for concrete with and without polymers.

4.4 Compressive strength

All concretes were designed for a compressive strength higher than 50 MPa. The results presented in Figures 13-15 show that adding polymers lead to strength increase between 2-15%, except for 5%TPE and 5%N-228, as it did not show an improvement at 28 days. When compared to the control samples, the incorporation of 3% polymer (granular and powder) has enhanced the strength of concrete by approximately 5% for PCL and Polymorph and 10-15% for powdered polymer including LDPE/EVA, GPE and TPE at 28 days. 3% polymer replacement seems to benefit the aggregates grain size distritution and bonding characteristics within the concrete matrix, as demonstrated by SEM investigation (Figure 10). This emphasises that there is a good bonding strength between the cement paste and the polymers. The explanation for this improvement could be the PSD of the polymers as the powders are very small and ≥ 1.18 mm, filling the gaps and consequently produce a stronger and denser concrete. Another factor could be the curing condition of the samples as after demoulding the samples were cured in warm water (40 ˚C) for seven days which softened the granular polymers, enabling them to spread and fill the voids. This could aid the shape (surface texture) change, which might create a better bond between cement paste and polymers.

Control mixture at 14 and 28 days present similar compressive strength. Similar trend was observed for PCL (granular, 3%), GPE and TPE (both 5%, powder).

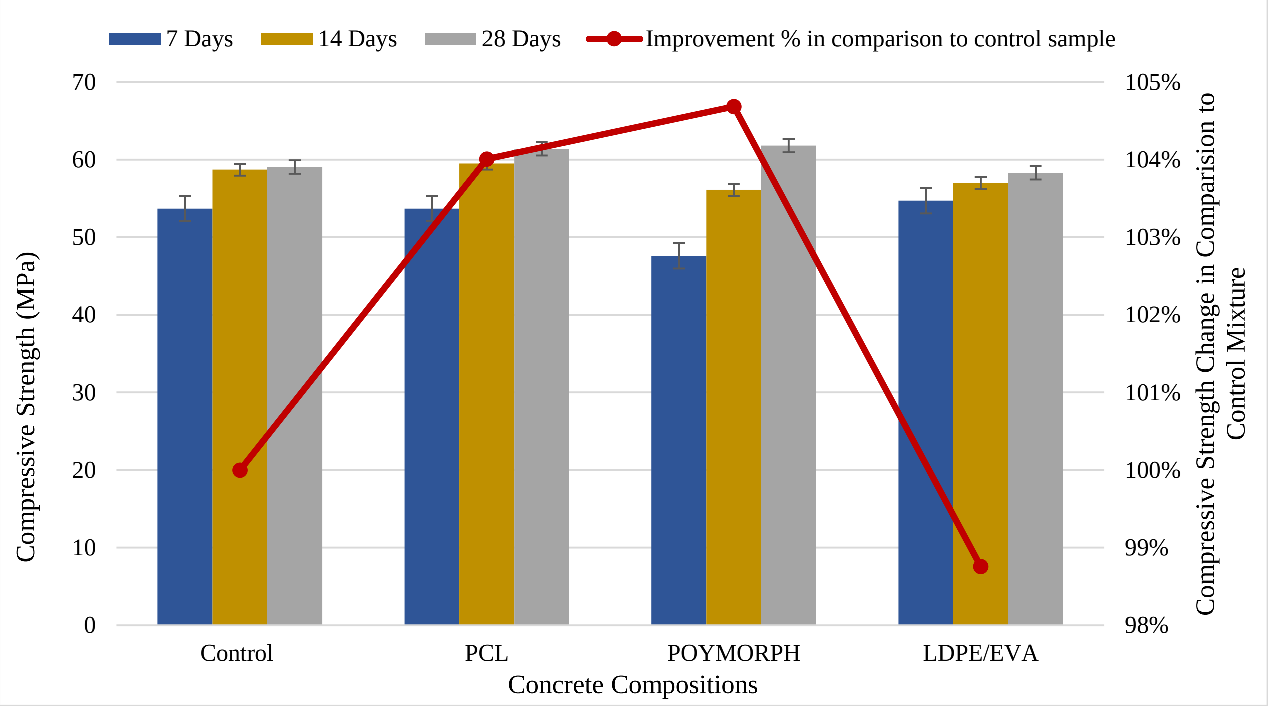


Figure 13 – Fine aggregate replacement by 3% granular thermoplastic polymer.

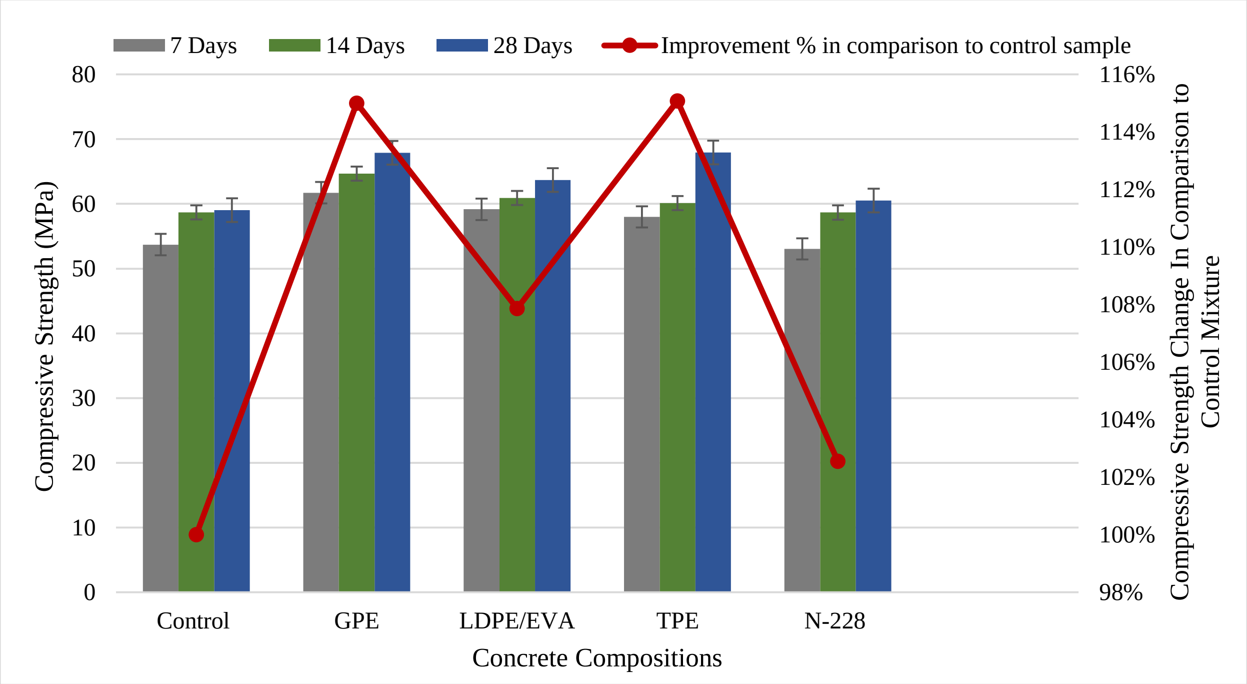


Figure 14 – Fine aggregate replacement by 3% powder thermoplastic polymer.

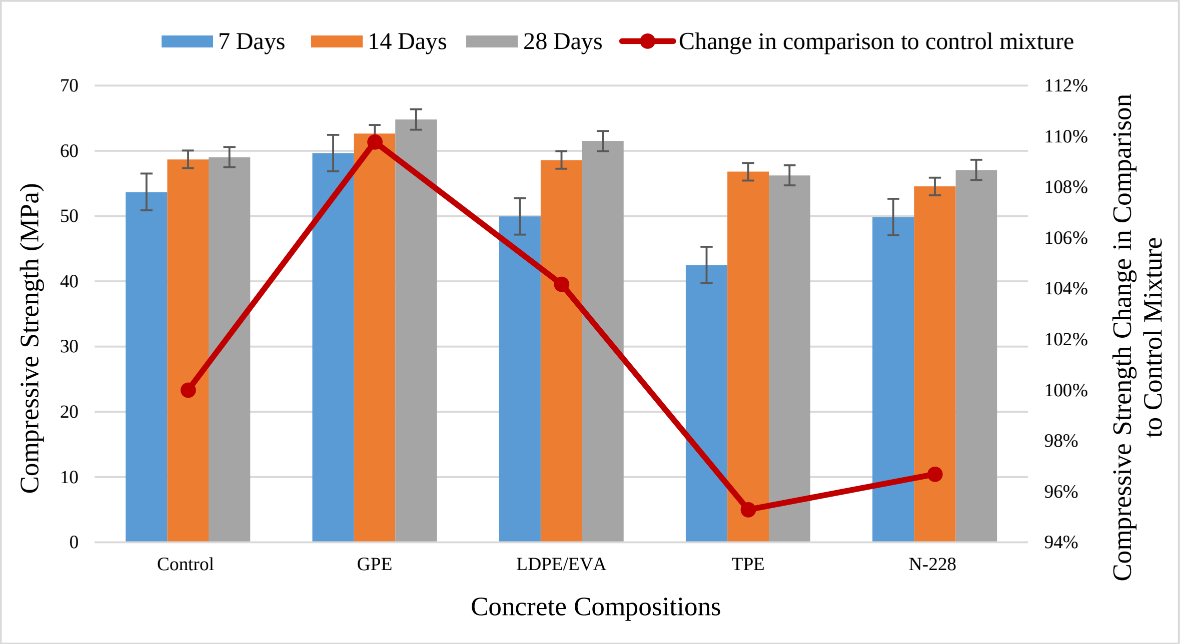


Figure 15 – Fine aggregate replacement by 5% powder thermoplastic polymer

4.5 Water absorption via capillarity

Figures 16 and 17 show the experimental findings of water absorption via capillary for control samples as well as concrete containing polymers for the first 2 and 28 days.

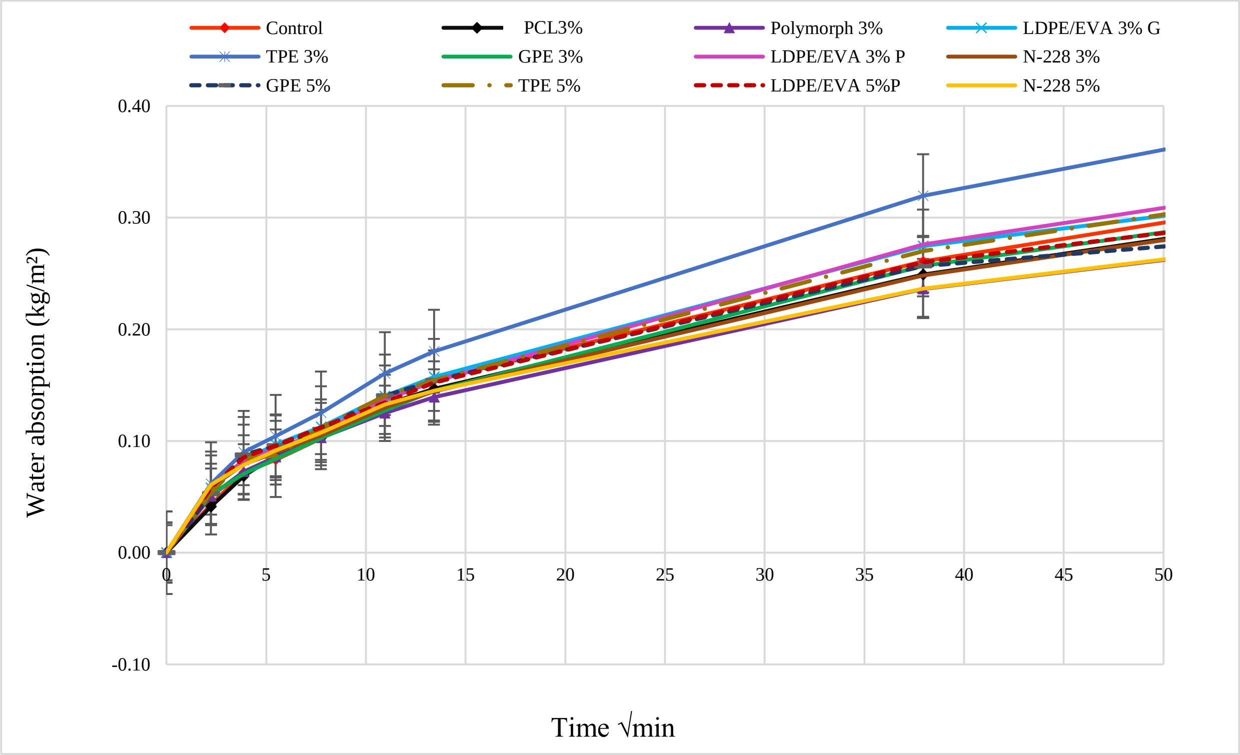


Figure 16 – Concrete water absorption via capillary for the first 2 days.

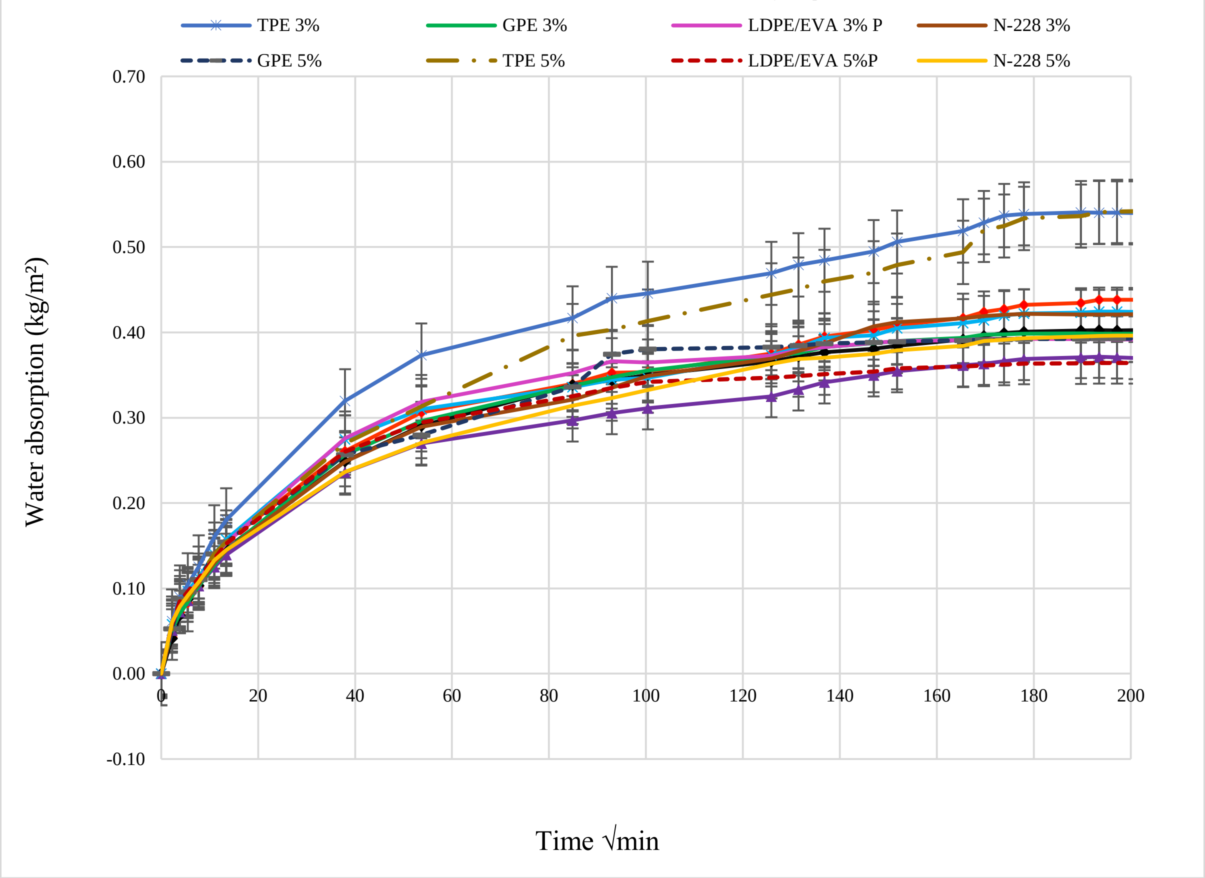


Figure 17 – Concrete water absorption via capillary for 28 days of duration.

In the first 2 days (figure 16) the majority of compositions lead to same water absorption rate via capillary except for TPE 3%, where the concrete composition seems to increase the rate of water absorption. However, at 28 days (figure 17) the capillary water absorption decreased for six types of polymers modified concrete, when compared to the concrete control sample. Polymorph 3% and LDPE/EVA 5% seems to be the ones that more effectively prevent the water migration, in comparison to control mixtures.

For the majority of polymers, their effect on preventing water migration via capillary seems to happen 2-3 days after the beginning of capillary test. This reduction is due to the hydrophobic nature of polymers and also, the capillary gaps that are blocked (filled by polymers) when the polymers are dispersed in the cement paste. Furthermore, the bonding strength between the cement paste and the polymers typically affects the concrete permeability, the stronger bond the less permeable is concrete (see figure 10). The concrete permeability reduced with the introduction of polymers and the stronger bonds decreased the gaps between cement paste and polymers, therefore, this type of concrete is able to protect steel from corrosion. The present study outcomes are in agreement with the findings observed by Akcaozoglu et al [25] and Maezouk et al [26] that stated that the hydrous transfer into the samples were much more slower with PET inclusion. As a result, the lowest degree of water penetration is achieved by 3% Polymorph and 5% LDPE/EVA content which is demonstrated in figure 18. According to [42]. LDPE granular or powder at 3% of incorporation leads to similar water absorption results.

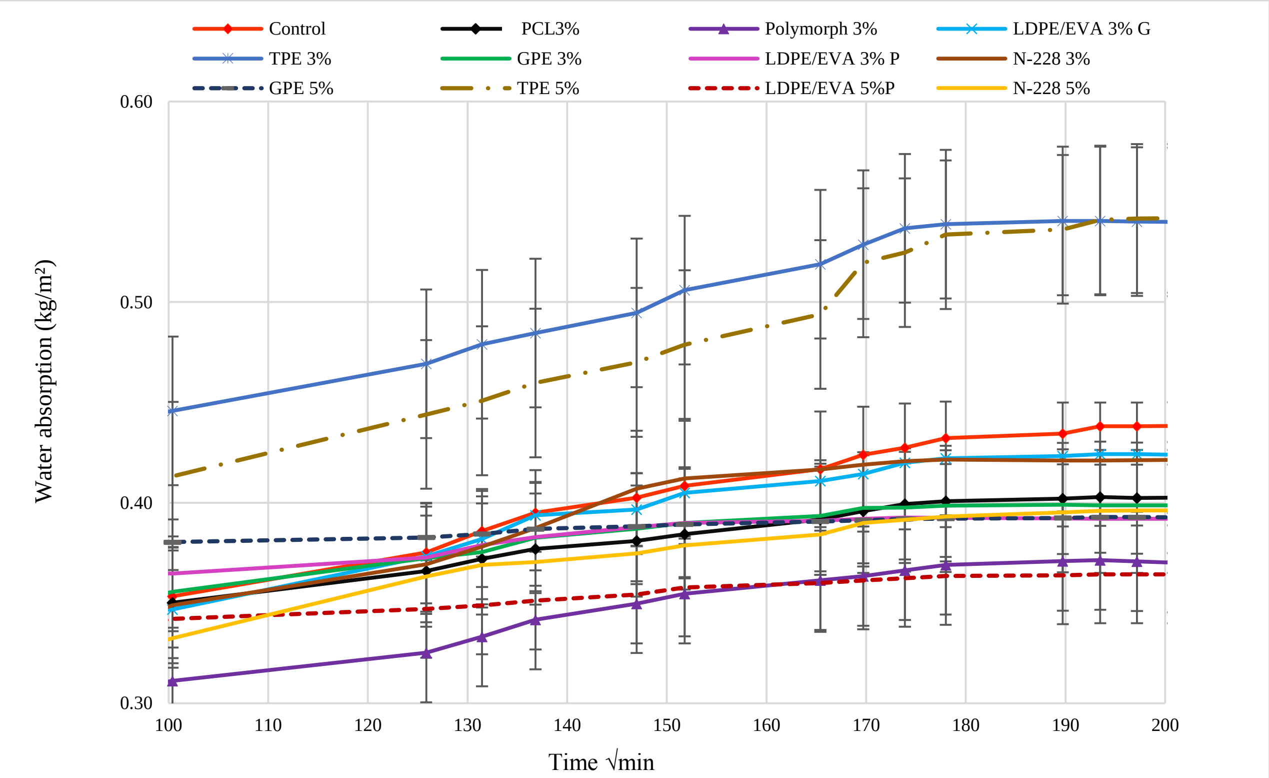


Figure 18 – Concrete water absorption via capillary from 7 to 28 days of testing.

However, the 3 and 5% TPE substitution resulted in higher water absorption, the main reason for this can be due to the shape and absorption properties of TPE which could be responsible for this behaviour. To confirm the experimental outcomes, concrete containing TPE powder was investigated under the SEM imaging in order to examine the morphology of the interfacial transition zone (ITZ) between the cementitious matrix and polymers. According to figure 10 (m&n), It was found that TPE is a porous polymer and increased the water permeability and microcracks within the concrete samples. From the solubility analysis it was found that the water can get inside the voids of TPE, but it cannot escape through it. This could explain the capillary constant value for 3 and 5% of TPE in figure 18.

4.6 Open porosity of concrete containing polymers

The water absorption provides an indication of the porosity level for the materials by determining the percentages for water absorbed under particular conditions [43]. Porosity is indirectly reflected by the water absorption characteristics such as the permeable connectivity of the pore [44]. The following figure shows the open porosity of concrete with and without polymers.

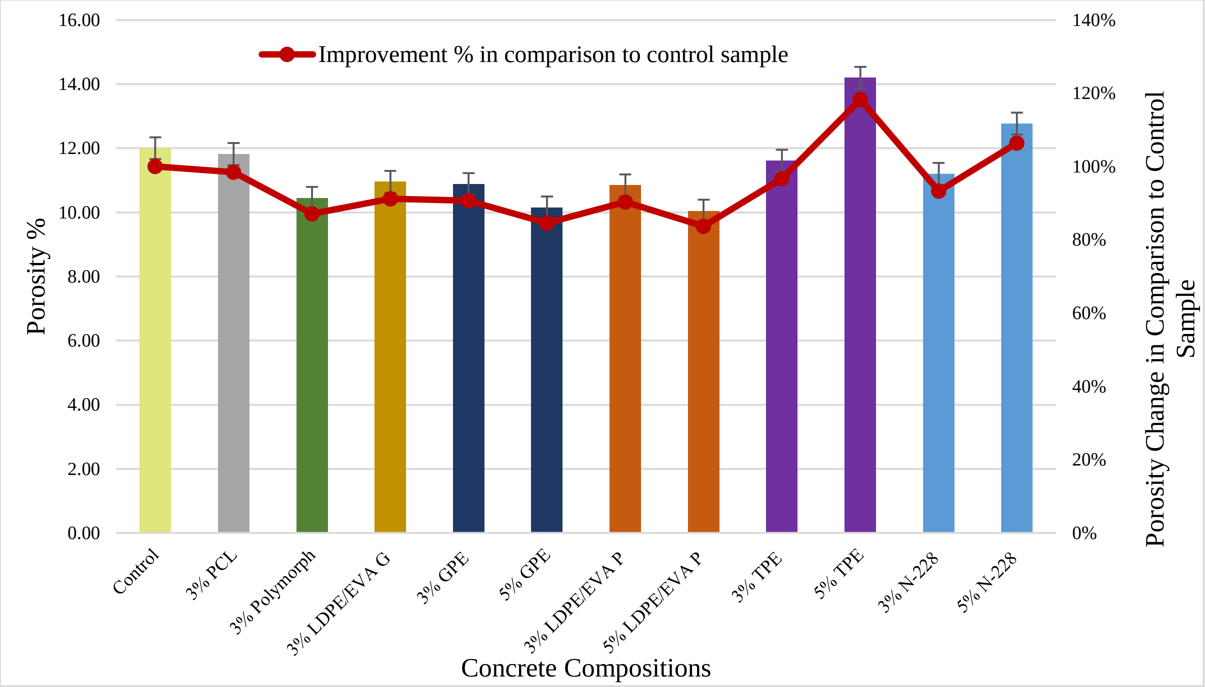


Figure 19 – Open porosity values for concrete with and without polymers.

Open porosity values seems to be very similar for all the compositions, whereas polymers tend to slightly decrease it by 2% from 12%. 5% TPE composition shows the highest porosity value. Obtaining the higher porosity value for TPE can be explained by its shape, as it was observed under SEM imaging that TPE is porous polymer and water can get inside the pores. The porosity reduced with the addition of 6 types of polymers, except for the TPE type of polymer, this improvement could be due to the shape and PSD of the used polymers. As Albano et al [21] reported that the flaky, coarse plastic aggregate resulted in a greater increase of the capacities of water absorption than the pellets shaped and fine plastic aggregates did. The stronger the bond between the plastic aggregate and cement paste, the lower will be the gaps between the polymers and the cement paste. The observed reduction of voids (gaps) between polymers and the cement paste, together with SEM results enphasise a good adhesive strength observed between polymer and the cement paste . With reducing the voids, the pore number decreases and the pore structure within the concrete becomes discontinuous and inefficient in the transport of liquids (water), resulting in decreased permeability.

4.7 Chloride migration

Figure 20 presents the chloride migration coefficient for the control sample and concrete containing polymers, according to NT BUILD 492 standard.

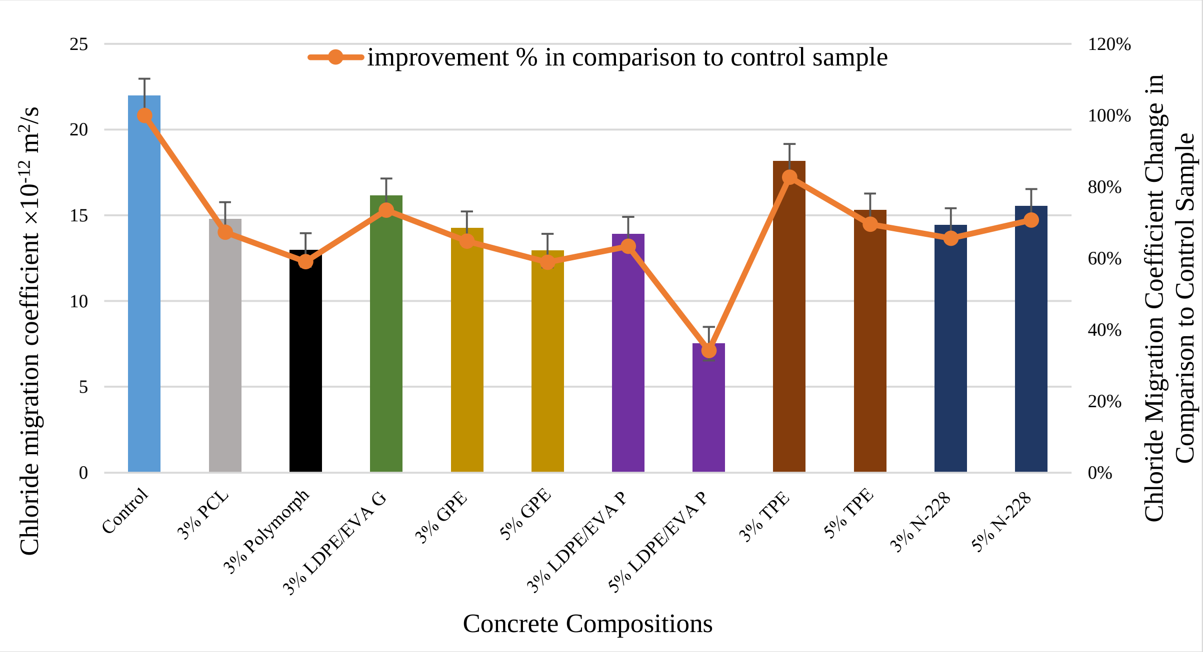


Figure 20 – Coefficient of migration of all concrete compositions.

Figure 20 demonstrates that the substitution of natural fine aggregates with polymers did significantly lead to a reduction of chloride ion transfer into the concrete samples, which is associated with an increase of the initiation time of corrosion. Also, chloride permeability may also be related to the open pores in terms of the amount of the fluid permeating through the pores of the concrete.

The fact that porosity (fig. 19) seems similar to all concrete compositions and the fact that capillary (fig. 17) and chloride migration coefficient are different, suggest that there are chemical reactions happening at the microstructure level, blocking the migration of ions, contributing to decrease the chlorides migration. The migration coefficient (D) improved when TPE or LDPE content increased from 3 to 5%. At 28 days, the concrete chloride migration coefficient decreases if TPE increases from 3 to 5%. This indicates that there is a chemical bonding of TPE to stop free chlorides migration inside concrete, which leads to a decrease of the coefficient of migration. This is possibly due to its molecular structure as shown in (table 1). TPE is more water repellent than the other candidate polymers because of having benzene in the molecular structure. For 3% of LDPE, it is evident here the benefit of the powder instead of granular polymer.

LDPE/EVA P also shows a chemical bounding with free chlorides from 3 to 5%. Kou et al [14] and Fraj et al [28] also observed a lower chloride diffusion coefficient of concrete containing polymers, which is in agreement with our results. 5% LDPE/EVA powder has recorded the maximum chloride migration coefficient improvement by 64% when compared to control sample. The minimum improvement is achieved by 3% TPE content, leading to a reduction by 17% in comparison to control concrete (see Figure 20).

Figure 21 – Chloride penetration in the concrete samples after 24 hours.

The direct replacement of 3% and 5% polymers has considerable properties in terms of mechanical and durability. Figure 21 emphasises the benefit of polymers impregnation and presents a comparison on the performance of polymer impregnated concrete related to the control sample. Figure 22 presents the impact of the use of polymers in concrete in comparison to OPC at 28 days.

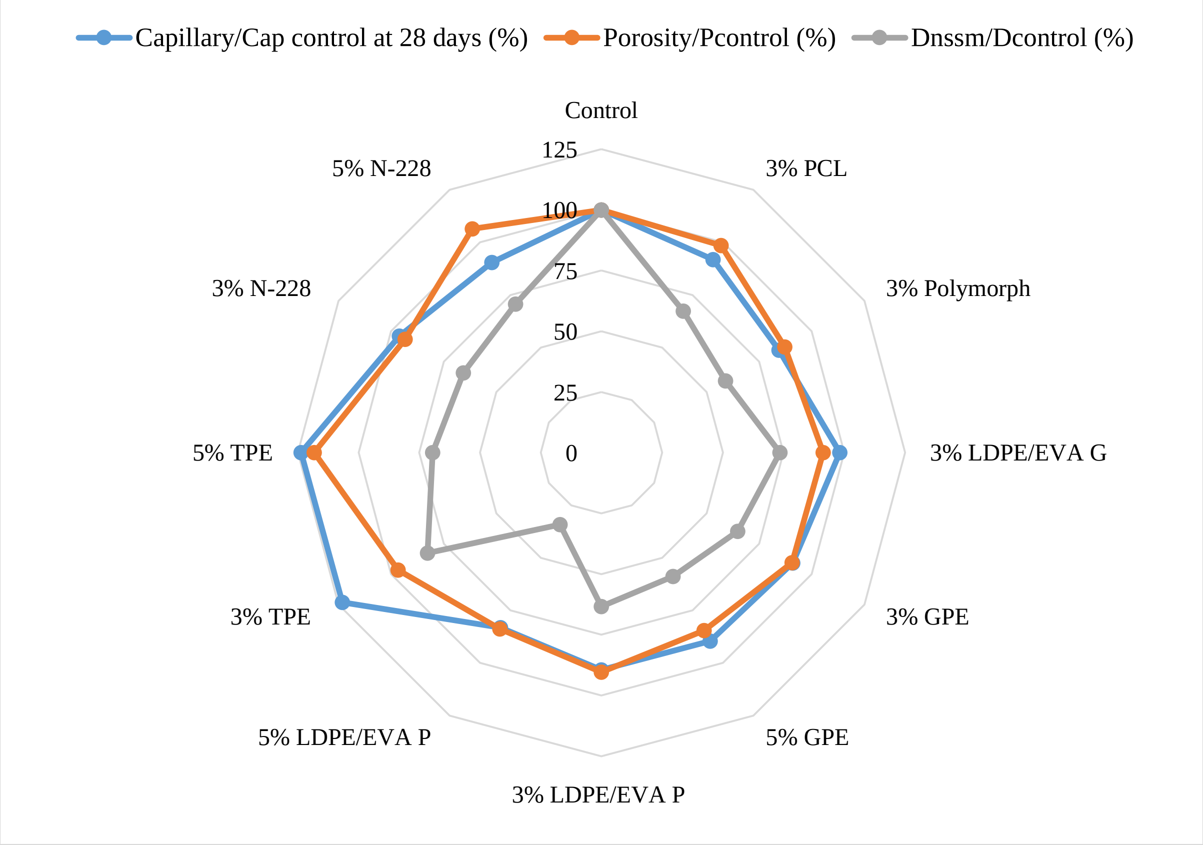


Figure 22 – Impact of the use of polymers in concrete in comparison to OPC at 28 days.

According to the previous figure, the polymers with better performance from a durability perspective are: Polymorph, GPE and LDPE/EVA powder/granular.

Polymorph and PCL are biodegradable polymers having a low melting point of 62 ˚C and softening point of 40 ˚C with an average molecular weight of 80.000 Da. Naturally, the smaller the molecular weight the shorter chain. Polymers with shorter chains have more end groups, which increases molecular mobility and free volume, making it easier for polymer chains to react to stresses. As a result, it is easier to block the pores and improve the durability of concrete.

However, because GPE is in powder form, it can easily block the pores within the concrete due to its size. Chlorides are carried by water. If water is unable to reach the interior of concrete, neither will chloride. Hence, chlorides do not travel on their own; they must be dissolved by water,, it's hard for concentrated iron solution to pass through the small pores of concrete.

The inclusion of LDPE/EVA into the concrete showed the best chloride migration coefficient value. This might be due to the modification of LDPE with EVA copolymer, resulting in an enhancement of the material toughness due to the increase of the adhesive strength. It is well known that EVA has been used in the modification of PE for better flexibility, toughness, and resistance to environmental stress cracking. Figure 22 highlights that the 5% LDPE/EVA P brings a great benefit to concrete from a durability perspective.

5. Conclusions

This research study the performace of seven different types of recyclable thermoplastic polymers for modified concrete at percentages of 3 and 5% of sand replacement, from a durability point of view. The following conclusions can be drawn from the findings of this experimental research.

1. The overall concrete workability improved due to the incorporation of the polymers. Polymer impregnation tends to increase concrete workability, making the concrete suitable for normal reinforced concrete manually compacted and heavily reinforced sections with vibrations. The workability enhancement is due to the particle size, shape and smoother surface texture of the used polymers when compared to the natural fine aggregate.

2. The incorporation of 3% polymer granular and powder enhanced the strength of concrete, approximately 5% for PCL and Polymorph and 10 to15% for powder polymer

including LDPE/EVA, GPE and TPE at 28 days when compared to the control samples. However, the strength improvement was less when the replacement level increased to 5%. SEM analysis proved that there is a good bonding strength between the cement paste and the candidate polymers except TPE type of polymer. The good bond is responsible for compressive strength improvement.

3. The porosity and water absorption via capillary of concrete decreased when introducing six different types of candidate polymers in comparison to concrete samples. The lowest degree of water penetration is achieved by 3% Polymorph and 5% LDPE/EVA content. The TPE substitution resulted in higher porosity and water penetration, the main reason was be due to the shape of TPE is a porous type polymer and increased water permeability and microcracks within the concrete samples, this has been investigated under SEM analysis.

4. The substitution of natural fine aggregates with polymers did significantly involve in reduction of chloride ion transfer into the concrete samples. At 28 days, the concrete chloride migration coefficient decreases if TPE increases from 3 to 5%, but porosity increases if 5% of TPE is added instead of 3%. For GPE the behaviour is the same either for 3 or 5%. This indicates that probably there is a chemical bonding of TPE to stop free chlorides migration inside concrete, which leads to a decrease of the coefficient of migration. The inclusion of LDPE/EVA into the concrete showed the best chloride migration coefficient value, leading to a reduction of 64% in comparison to control concrete mixtures. This might be due to the modification of LDPE with EVA copolymer, resulting in an enhancement of the material toughness due to the increase of the adhesive strength.

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