Chapitre 3 : Caractérisation thermomécanique de bétons faits avec des agrégats de plastique post-consommation

Résumé

Les déchets de plastique constituent un enjeu environnemental majeur, et des techniques de recyclage innovantes doivent être développées pour augmenter les taux de récupération et de recyclage de façon significative. Ce chapitre présente des travaux qui visent à complémenter les études réalisées dans le domaine du béton incorporant des agrégats de plastiques recyclés, en utilisant une variété de plastiques en provenance de flux de matières d'un centre de tri. Trois séries expérimentales de béton ont été caractérisées, en portant une attention particulière sur les propriétés améliorées par la présence d'agrégats de plastique, soit la résistance post-pic et la conductivité thermique. Le volume d'agrégat de plastique dans les recettes de béton, les propriétés physico-chimiques des différents plastiques utilisés, et leur variation entre plusieurs échantillons récupérés sur une année complète étaient les variables à l'étude. Des tableaux de corrélation de Pearson ont été utilisés pour identifier les variables ayant le plus d'influence sur les propriétés du béton. Les recettes développées les plus performantes ont démontré des ductilités élastiques égales ou supérieures au béton témoin (sans agrégats de plastique), tout en affichant une diminution de module élastique de 10-12% (les agrégats de plastique représentaient 10% du volume des agrégats fins). La conductivité thermique était directement reliée au volume d'agrégats de plastique et d'air entraîné dans le béton, tandis que les propriétés mécaniques étaient surtout influencées par les caractéristiques des agrégats, comme la géométrie et la densité brute. Cette étude démontre que cette technique de recyclage du plastique peut être une alternative intéressante aux coûteux procédés de tri et conditionnement du plastique, mais qu'un contrôle qualité de certains paramètres clés demeure essentiel pour stabiliser les performances obtenues sur le produit fini.

Characterization of Concrete Composites with Recycled Plastic Aggregates from Postconsumer Material Streams

Abstract

Plastic waste is a major environmental issue and innovative recycling techniques are needed to boost its recovery rates and value. This research aims to complement previous studies in the field of concrete made with recycled plastic aggregates (PAG) by using a variety of plastics from mixed material streams at a materials recovery facility. Three series of experimental mixtures are characterized with emphasis on concrete post-cracking strength and thermal conductivities, where PAG may show beneficial effects. The effects of PAG volume in the mix, PAG physical properties and temporal variations in material streams are discussed. Pearson correlation tables are used to identify properties of importance and their effects on concrete. Our best mix designs had elastic toughness values equal to or superior to the reference mix, while experiencing a drop of 10-12% in elastic modulus (PAG represented 10% of fine aggregates volume). Thermal conductivity is directly related to the volume of PAG and entrained air in concrete, since they are both much less conductive than mineral aggregates. Our study demonstrates that certain PAG properties, such as shape and bulk density, are important drivers of concrete mechanical performances. While careful monitoring of certain properties is essential, this recycling technique could be a promising alternative to expensive material sorting processes.

3.1 Introduction

The environmental pressure generated by the exponential production of plastics has reached a critical point. Worldwide plastics production was 299 million metric tons (MT) in 2013, with an average yearly growth rate of nearly 5% [2]. Despite persistent efforts by governments, businesses and individuals to increase plastic recovery and recycling rates, an estimated 4.8 to 12.7 million MT end up as debris in rivers and oceans [15], and global recycling rates remain low, even in regions with a developed waste management infrastructure. For example, postconsumer plastic recycling rates are below 40% in all European countries [2]. The province of Quebec (Canada) claims a household plastics recovery rate below 32%, but the actual recycling rate is expected to be even lower, as only 17% of the total was reported in transactions with recyclers [7, 10]. Postconsumer plastic waste is known to pose difficult challenges to recycling [17, 43]. The wide variety of polymer resins found in municipal recyclable streams, their general incompatibility when molten for mechanical recycling, and high levels of impurities create a context where extensive sorting, conditioning and upgrading processes are required [21]. Plastics are generally sorted by polymer type in material recovery facilities (MRF), either manually (based on packaging visual appearance) of mechanically with optical sorting units [16, 26, 44]. Sorted streams of sufficient volume are then granulated, washed and often pelletized by conditioners to obtain an easily marketable commodity [45]. Often, economic reasons prevent the successful implementation of this value chain, although recent technological advances have lowered processing costs [43]. On the other hand, door-to-door collection programs of mixed recyclable materials are generating ever increasing volumes of plastic available for valorization. There is therefore a clear need for recycling technologies that can process large volumes of plastic at low cost (i.e. with minimal prior conditioning). In addition, these techniques could be considered as an alternative to landfilling or incineration for plastic streams that are severely contaminated, such as marine debris or municipal solid waste.

Concrete is the most employed construction material today reaching presently about 10 km^3/yr, which is about 5, 7.7 and 10 times more than the amount yearly employed for fired clay, timber, and construction steel, respectively [46]. There are numerous opportunities for improving the ecological impacts of this massively employed material [47]. Concrete represents an important outlet for the valorization of large volumes of waste as replacement for mined resources. For instance, construction waste is already used to produce lightweight concrete aggregates for structural use, thus facilitating a closed-loop system where waste generators and users are geographically close [46, 48]. This approach helps tackle two major environmental issues: (i) the ever-increasing amount of anthropogenic waste in landfills and the environment; (ii) the concerns associated with extracting non-renewable natural resources. In such context, the idea of using plastic waste as a raw material for concrete lends itself to the trend of urban mining, which is described as the “systematic reuse of anthropogenic materials from urban areas” [49].

Numerous research on recycled plastic aggregates (PAG) used as sand replacement in concrete mixtures have been undertaken, and all unanimously stress the ecological potential of the technology as the primary driver for the development of this field [35, 50, 51]. Interestingly, many have also demonstrated a potentially beneficial effect of PAG on some physico-mechanical concrete properties such as ductility, flexural toughness, density and thermal resistance [52–54]. Table 3.1 summarizes the results of recent studies available in the open literature. To put the following discussion into context, Table 3.2 reports typical Young’s modulus (E), tensile strength (f_t) and thermal conductivity (λ) of the polymers considered in this study along with typical concrete raw materials such as aggregates, sand and cement paste.

The introduction of PAG usually negatively affects mechanical properties, for instance, E and f_c. It has been reported that E decreases proportionally to PAG volume [35]. Correira et al. [55] found that E decreased by 13-31% for mixtures where sand was substituted by polyethylene terephthalate (PET) aggregates at 7.5% volume. This was expected as PET aggregates have a much lower E than typical mineral aggregates (Table 3.2). It is also worth noting that a variation in the results was due to varying amounts of water used in the experimental mixtures to keep a constant slump. Indeed coarser, more angular PAG tend to decrease fresh concrete workability as opposed to rounder sand particles or pellet-shaped PAG [55]. The authors observed, in agreement with previous studies (Albano et al. [35] and Saikia et al. [56]), that PAG size can significantly impact fresh concrete workability, and thus porosity, compaction and mechanical performances in hardened concrete.

For f_c, previous studies have found that replacing sand by PAG also leads to a noteworthy strength loss. Hannawi et al. [57] reported a decrease in f_c of respectively 30% and 28% by replacing 10% of fine aggregate volume with PET and polycarbonate (PC) aggregates.

The loss of f_c was well correlated with the loss of E, suggesting similar underlying causes. Visual inspections of the interface zones under an electron microscope showed large gaps around hydrophobic PAG, which suggests a poor bonding with the cement paste [57–59]. The interface zones between PAG and the surrounding cement matrix is apparently quite porous, while the cement-mineral aggregates interface exhibits better bonding [57].

Table 3.1 : Summary of the reported effects of PAG on key concrete properties.

Polymer used for PAG	Size[1]	PAG volume[2]	f_c loss[3] (%)	E loss[3] (%)	λ loss[4] (%)	Slump (mm)	Reference	Notes
PET	< 8 mm	7.5	31	31		116	[55]	e
	< 4 mm	7.5	14	13		127		
RPOMIX (compound)	< 2 mm	10	48.5				[54]	a
		20	75.7		50			
PET	< 10 mm	10	30.5				[57]	e
		50	69	68.4				
PC	< 5 mm	10	27.2				[57]	e
		50	63.9	61.9				
PET	< 11.4 mm	20	49	48		0	[56]	e
	< 2.6 mm	20	38	45		45		
PVC	< 5 mm	15	18.6	13.8			[53]	b
		30	21.8	18.9				
PU	< 4 mm	13.1	56		55	80	[60]	c
		33.7	94		85	30		
PET	< 2.5 mm	10	0		10.3		[61]	d
		30	0		20.6			
		100	86		73.8			

[1] Estimated from a 95% passing value from size analysis curves; [2] Expressed as % of fine aggregates in concrete, and as % total volume in mortars; [3] Under wet curing conditions, when specified; [4] Dried specimens; [a] Hydraulic lime mortars; [b] Lightweight concrete with expanded clay aggregates; [c] PUR foam lightweight mortars; [d] Mortars; [e] Concrete

Table 3.2 : Some properties of high volume polymers and concrete materials.

Material	E (GPa) [29, 31]	f_t (MPa) [29]	λ [W/ (mK)] [30, 31]
PET	2.1-3.1	55-80	0.15
PE	0.6-1.4	18-30	0.33-0.52
PVC	2.7-3.0	50-60	0.17-0.21
PP	1.3-1.8	25-40	0.12
PS	3.1-3.3	30-55	0.105
Quartzite sand	70	-	4.45
Limestone gravel	70	-	2.29-2.78
Cement paste (w/c=0.5)	36-40	-	1

The post-cracking strength (PCS) of concrete is an important property to avoid brittle failure in concrete structures and help dissipate high energy, for example in the case of a seismic event [64]. Flexural tests are often used to characterize the toughness (also called fracture energy) for concrete and mortars incorporating PAG [35, 57, 65]. The addition of PAG in concrete was found to decrease the strength, but to enhance the fracture energy, which is usually calculated as the area under the stress-strain curve. Hannawi et al. [57] reported slightly lower f_t in mortar mixtures incorporating PET or PC as fine aggregate, but up to 6 times higher fracture energies when compared to a conventional sand and cement mortar. These results suggest that ductile PAG inclusions delay crack propagation [57]. As f_t and f_c seem to vary together with the use of PAG, the same could be observed in flexural and compressive PCS, although the later has not been investigated in previous research. Compressive PCS of concrete has several beneficial effects on concrete structures, such as: better confinement to the longitudinal reinforcement, reduced amount of steel stirrups, lower risk of instability in the steel reinforcement, and enhanced rotational capacity of a beam section [64].

For thermal properties, PAG could impede heat flow in concrete thanks to the low λ of polymers [61, 66, 67] (see Table 3.2). For instance, Marzouk et al. (2007) found that the conductivity of mortars went from 1.26 down to 1.13 and 0.63 W/ (mK) by replacing fine aggregates with 10% and 50% PET aggregate, respectively [61]. This is particularly promising for enhancing the insulation of buildings applications as PAG could not only help insulate concrete, but also be used to control the heat transfer properties of thermal mass components in energy efficient buildings [67].

The effect of PAG on concrete durability has not been fully studied. Kou et al. [53] found that polyvinyl chloride (PVC) aggregates significantly reduced the drying shrinkage and chloride ion penetration of concrete. Those effects are particularly important for reducing the risk of cracking in concrete and enhancing structural durability.

The works cited above show that PAG can significantly alter the physico-chemical properties of concrete mixtures, even in relatively low volumes. From a waste management perspective, it is well known that recycled material streams are prone to variations in their physico-chemical properties due to factors such as time, industrial processes and sourcing of material [31]. Although it is of practical importance, in-depth investigations of different polymers properties, and the effects of sampling bias of postconsumer material streams, on the overall properties of concrete have not been reported in previous studies. Hannawi et al. [57] used both PET and PC aggregates in the same mix designs and reported differences in their properties (Table 3.1), which calls for further investigations. In this regard, quality control specifications on raw plastic material properties need to be developed to limit the uncertainty in the desired performances of the resulting PAG concrete composites.

The combination of a high PCS and a low λ may make the upcycling of postconsumer plastics as aggregate replacement in concrete a promising application. By considering plastic collected from municipal waste material streams, the scope of this research is twofold: (i) study the effect of PAG on common concrete properties, with the addition of PCS and λ; (ii) consider the effect of polymer composition, and intrinsic fluctuations in sorted postconsumer plastic streams, on concrete properties.

3.2 Materials and methods

3.2.1 Cementitious materials

Quarry sand with a maximum size of 4 mm and limestone aggregate of 2.5-10 mm were provided by the Center for Research in Concrete Infrastructures of Université Laval. The size distributions of sand and limestone aggregates are shown in Figure 3.3. A Portland cement GUP type (Lafarge, St. Constant) was used. An air-reducing agent (Eucon Air-Out, Euclid Canada) was used to control the volume of entrained air in concrete samples. Mix designs are listed in Table 3.4.

3.2.2 Plastic material

A total of 434 kg of mixed postconsumer plastics were collected from a MRF (Gaudreau Environment Inc., Victoriaville, Canada). The sampling was done in 7 samples of about 60 kg from March 2015 to February 2016 (1 sample in March, 2 samples each in April, October and February). The samples were labelled #1 to #7 in chronological order of sampling. The material consisted primarily of mixed, rigid plastic packaging for food and household items that had been manually sorted from a commingled recyclable materials stream. Then, the collected samples were further sorted by running their contents under a near infrared optical sorting unit (Eagle Vizion, Sherbrooke) programmed to eject plastics based on a different polymer type (in order: Polypropylene (PP), Polyethylene (PE), Polystyrene (PS), Polyvinyl chloride (PVC)). The process is illustrated in Figure 3.1 and polymer composition in % weight of the plastic samples is given in Figure 3.2. Each of the 35 ejected fractions was manually inspected for contaminants, and a NIR spectrometer (Eagle Vizion, Sherbrooke OEM) was used to identify polymers. Residual material from the samples (“Others”) consisted of mixed plastic items that could not be ejected by the machinery due to size or shape, as well as all other polymer types such as PET, Polylactic acid (PLA), PC, polymer blends (such as multi-laminated plastics) and unidentifiable materials. Loose non-polymeric waste such as paper or glass was discarded.

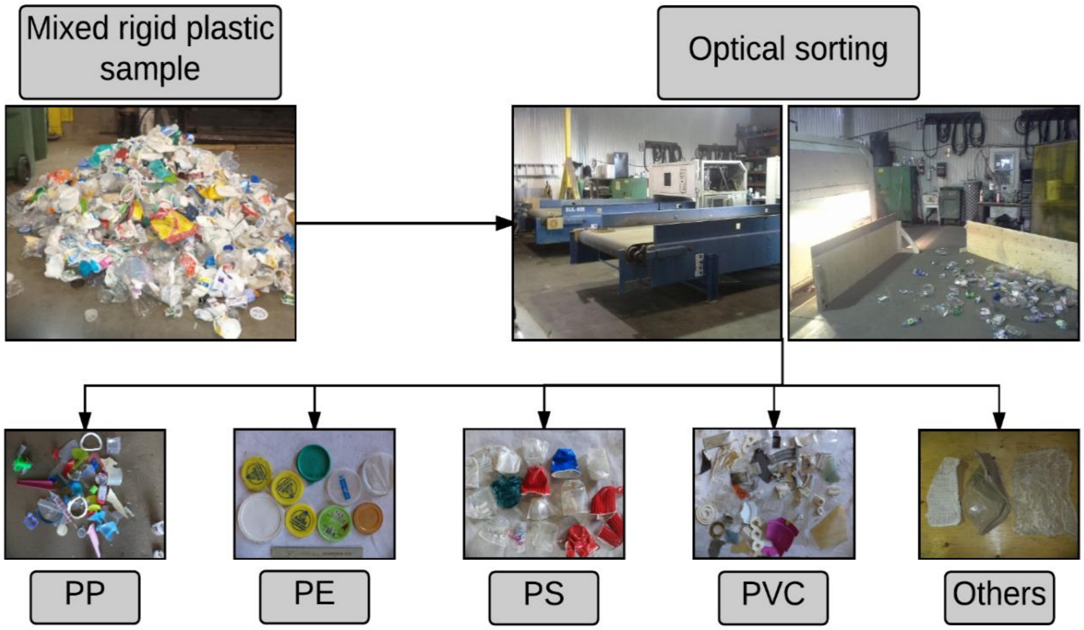

Figure 3.1 : Separating plastic samples into polymeric fractions.

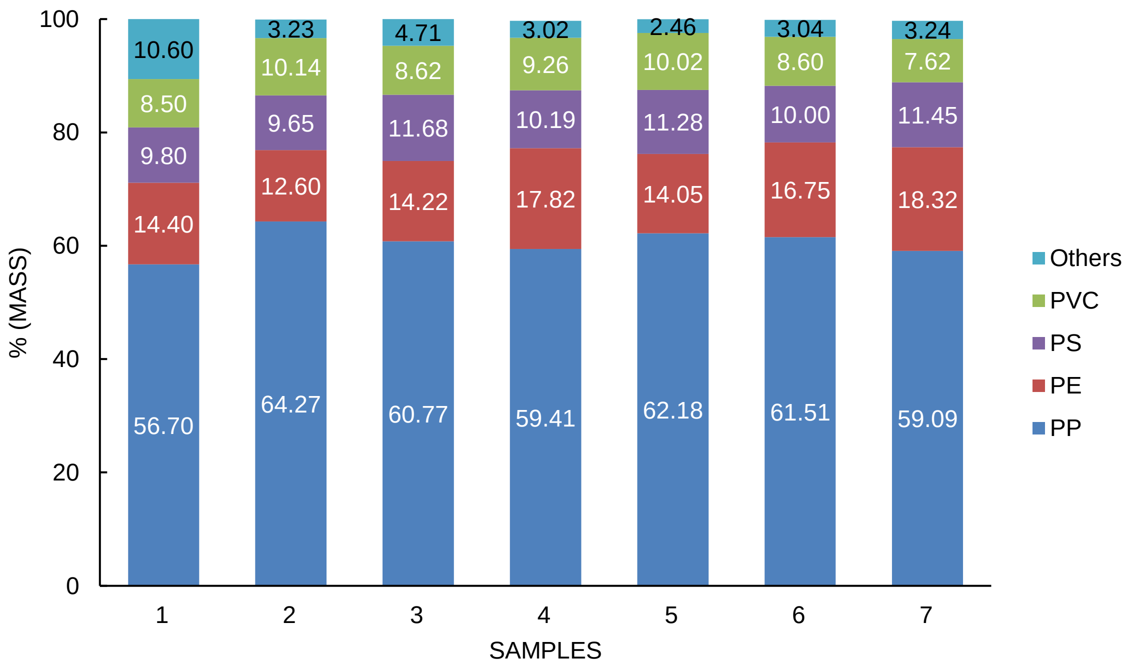

Figure 3.2 : Polymer composition of mixed plastics collected at the MRF vs. sample number.

3.2.3 Plastic aggregates (PAG)

Some of the fractions were selected to produce PAG and were specifically labelled, as shown in Table 3.3. Fractions that consist mainly of one polymer type were randomly selected among mixed samples #1-#7 and labeled with the acronym of the polymer. The MIX label indicates that all sorted fractions for a given sample were recombined into a mixed sample. Fractions that are labelled by a number (PS-PVC 1 to 5) refer to a combination of the PS and PVC fractions of a given mixed plastic sample. PS and PVC have the highest E of all polymers (Table 3.2) and were therefore selected to hypothetically produce mixtures with better mechanical properties. At the same time, these polymers have the lowest value on current recycling markets and are good candidates for innovative recycling solutions.

Table 3.3 : Fractions selected for this study.

Mixed stream sample ID	Selected polymer fractions	Mixtures of PS-PVC fractions
#1	MIX: (PP, PE, PS, PVC and Others)	-
#2	n.c.	n.c.
#3	PP	PS-PVC1
#4	-	PS-PVC2
#5	PE	PS-PVC3
#6	PVC	PS-PVC4
#7	PS	PS-PVC5

The above fractions were ground in a rotary knife granulator with a mesh size of 5 mm to generate “coarse” PAG. Fractions 1-5 were sieved at 1.7 mm to generate “fine” PAG. The plastic particles were passed through a stack of sieves arranged in decreasing size to determine their size distribution according to standard method ASTM C136 [68]. Finally, half of the sieved MIX fraction was recombined to generate a “graded” size distribution that matches a Fuller distribution in accordance with the requirements of standard CSA A23.1.14 for fine aggregates [69]. All obtained size distributions are shown in Figure 3.3.

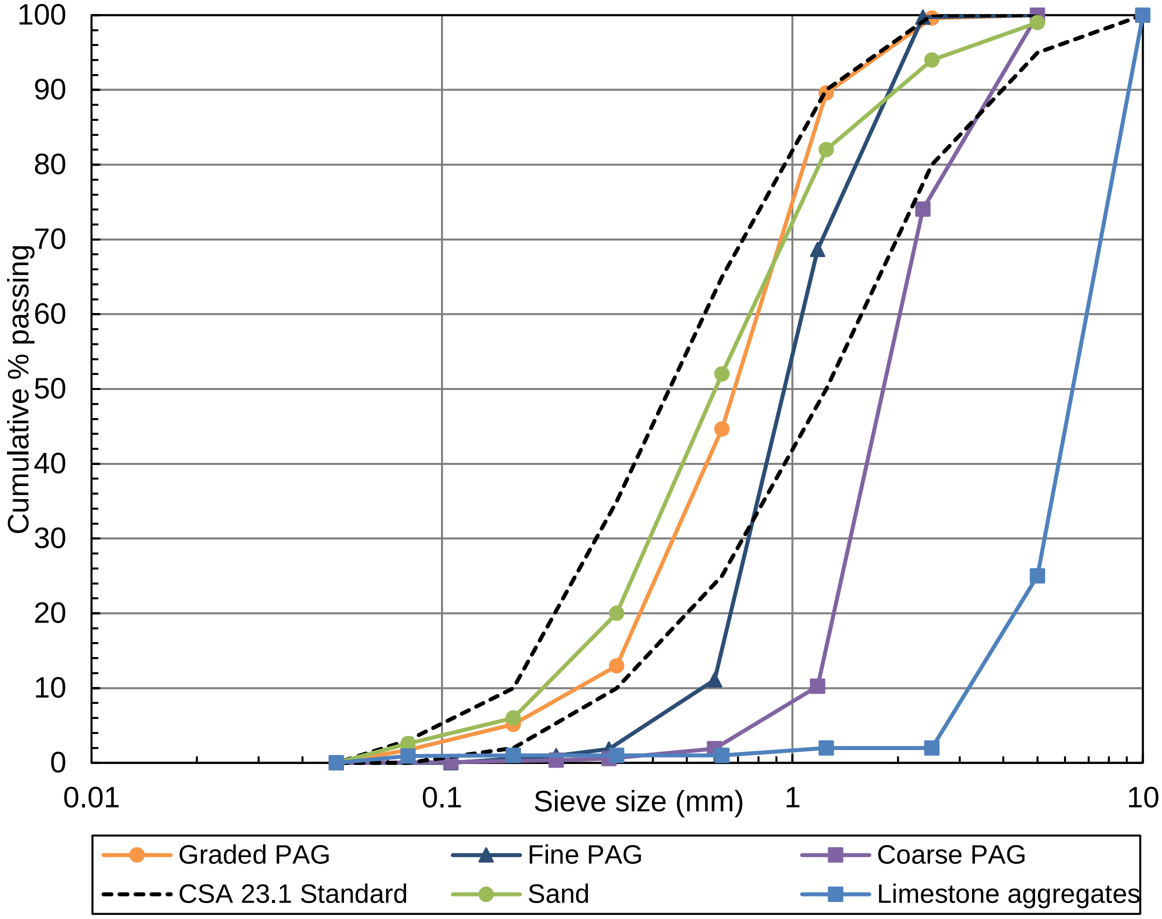

Figure 3.3 : Size grading analysis of the fine aggregates used in this study.

Un-compacted bulk densities of PAG were carefully estimated by means of a calibrated stainless steel vessel of known volume. The bulk density of aggregates can be used as a measure of angularity or particle shape, since randomly arranged particles that are closest in shape to a sphere will occupy the most volume in a given space [36]. It is important to consider particle shape when studying PAG as previous studies demonstrated their significant effect on fresh concrete workability, and a possible impact on mechanical performances. Furthermore, particle shape may vary strongly between plastics, notably because of different thicknesses in packaging items [26]. In this regard, a Shape Index was defined as follows, to represent the volume of interstitial spaces between un-compacted PAG:

$$\varphi = 1 - \frac{(m/\rho)}{V_{bulk}} \tag{1}$$

where φ is the % volume of voids between the uncompacted plastic particles, V_{bulk} is the vessel volume, m is the mass of PAG in the vessel and ρ is the experimentally determined plastic apparent density.

The PAG ρ was determined with representative samples of 30 g in three replicates, using a helium gas pycnometer (AccuPyc II 1340, Micromeritics) and standard procedure ASTM D5550 [39].

Thermogravimetric analysis (TGA) was performed in an oven (Q5000 TGA, TA Instruments) on homogenized PAG samples of 20-50 mg in three replicates, according to standard procedure ASTM E1131 for the compositional analysis of polymers [70]. Samples were heated to 600°C at a rate of 10°C/min in a N_2 inert atmosphere, then combusted in the presence of O_2 at a rate of 10°C/min to reach 750°C. Some TGA curves (mass vs. temperature) labeled with analysis points are shown in Annexe 2, and the measured parameters are reported in Table 3.4. At a given heating rate, anaerobic thermal degradation (pyrolysis) of a specific polymer occurs within known temperatures in the 200-600°C range. First, the derivative of mass vs. temperature TGA curves was used to identify several degradation steps, according to a method detailed by Ehrenstein et al. [40]. The largest continuous mass loss was associated with the polymer content in the sample, and used to calculate a "% Polymer" parameter. The experimental temperatures associated with this mass loss were corroborated with polymeric pyrolysis temperature ranges reported in the literature [41]. In the specific case of PVC, polymer pyrolysis occurs in two steps and generates carbon-based residual compounds (char) [40]. Therefore, it was necessary to combine mass losses for 2 pyrolysis steps and the mass loss immediately following combustion at 600°C to estimate the polymer content. The remaining mass after combustion consists of inorganic material from polymer additives, reinforcement material or surface contamination of the sample.

3.2.4 Mix designs and concrete samples

A normal concrete mix design with a compressive strength of approximately 40 MPa was chosen, which consisted of: 400 kg/m^3 cement; 1024 kg/m^3 limestone aggregate; 724 kg/m^3 quarry sand and 200 kg/m^3 water (% air = 3.7, slump = 125 mm). In the recycling mix design, a volumetric fraction of sand was replaced by PAG samples, while ensuring that final concrete volume remained the same for all mixtures. The water-to-cement ratio (w/c = 0.5) was kept constant in this study and no corrections for slump were attempted. Table 3.4 summarizes the mix designs of the 14 experimental mixtures, which can be classified in the following three series: (i) Series "a" consisted of 4 mix designs with the same plastic material (MIX fraction, which represents a mixed plastic material stream prior to sorting), while varying the PAG volume and size distribution; (ii) Series "b" consisted of 5 mix design which were made using the same sand replacement (20%) with PAG, but different plastic kinds; (iii) Series "c" consisted of 5 mix design with 10% sand replacement with PAG fractions PS-PVC 1 to 5 to capture variations in the plastic stream.

Table 3.4 : Mix-designs studied in this work.

Tests series	Fraction	Substitution rate [%]	PAG size distribution	Air reducing agent (mL/m^3)	Concrete ID
Series a	MIX	5	Coarse	0	a5
	MIX	10	Coarse	0	a10
	MIX	10	Graded	0	a10_graded
	MIX	20	Coarse	0	a20
Series b	PP	20	Coarse	50	bPP
	PE	20	Coarse	50	bPE
	PVC	20	Coarse	50	bPVC
	PS	20	Coarse	50	bPS
	MIX	20	Coarse	50	bMIX
Series c	PS-PVC 1	10	Fine	25	c1
	PS-PVC 2	10	Fine	25	c2
	PS-PVC 3	10	Fine	25	c3
	PS-PVC 4	10	Fine	25	c4
	PS-PVC 5	10	Fine	25	c5

Mixtures were cast according to CSA A23.22C and CSA A23.23C standard procedures [69]. Each mixture was cast into 6 cylinders (height of 150 mm and diameter of 75 mm) for mechanical testing. Additionally, certain mixtures were cast into 3 cylinders (height of 200 mm and diameter of 100 mm) for thermal conductivity, water absorption and density analyses. All cylinders were cured at 100% humidity for 28 days prior to mechanical testing, and 15 days for thermal testing. The air content and slump of fresh concrete were also characterised according to standard procedures CSA A23.24C and CSA A23.25C, respectively [69].

3.2.5 Concrete characterization

3.2.5.1 Elastic Modulus and Compressive Strength

A hydraulic press was employed to carry out compression testing. To measure the deformation during compressive testing, three linear variable differential transducers (LVDT) were mounted on the cylindrical samples by means of two aluminium rings at a distance of 100 mm. E was determined by averaging the slopes between the min. and max. loads of the last two out of three cycles in the elastic regime at a controlled rate of 1.100 kN/s. The stress-strain curve of each cylinder was obtained by imposing a strain of 0.25 mm/min. The test was stopped when the residual stress was equal to 30% of the maximum recorded (f_c). Results reported in section 3.3.3 are averages of 4-5 replicates.

3.2.5.2 Water absorption and concrete dry density

Dry densities of experimental concrete mixtures were determined by drying cured cylinders in an oven at 60°C until the recorded mass change was < 0.5% in 24 h. Dimensions of the cylinders were recorded to the nearest 10 µm and used to approximate their volumes. Water absorption was calculated after immersing the cylinders in boiling water for 5 h. Calculations and detailed procedures are found in standard CSA A23.2-11C [69].

3.2.5.3 Thermal conductivity

Thermal conductivities were obtained using oven-dried cylinders (average of 2 replicates) under thermal equilibrium conditions at an average temperature of 10°C. Each cylinder was fitted with two disc-shaped thermal flowmeters in a fully isolated chamber, and the power flux in W/m^2 was automatically acquired for a controlled temperature gradient set at 5°C between the top and bottom disks. A full description of the apparatus can be found in [71]. Thermal conductivity of the sample is given by:

$$\lambda = q * h/\Delta T = W/(mK) \qquad (2)$$

where *h* is the specimen height, q is the power flux and ΔT is the temperature gradient.

3.2.5.4 Toughness indices

The compressive stress-strain curve of each mix design was calculated from an average of 4 to 5 replicates for all experimental mixtures, and 10 replicates for the reference concrete. The end of linearity (EOL) point of a stress-strain curve was conventionally defined as the strain at which the curve tangent departs from E by more than 15%. Then, the area underneath the stress-strain curve up to the EOL strain was defined as the elastic toughness (W_e). Finally, two toughness indices, I_3 and I_5, were calculated by integrating the stress strain-curve to 3 and 5 times the EOL strain respectively, normalized by the elastic compression toughness, as follows:

$$I_3 = W_{3e} / W_e \tag{3}$$

$$I_5 = W_{5e} / W_e \tag{4}$$

3.3 Results and discussion

3.3.1 Properties of plastic aggregates

Optically sorted plastic samples were processed to PAG and characterized, and the results are shown in Table 3.5. The polymer concentration (% Polymer) obtained through TGA was used as a measure of the relative purity of the raw material. The concentration of impurities (volatiles, char or inorganic compounds) was found to be below 10% in all streams, and represented on average approximately 5% of sample weight. Contaminants in a plastic stream may include paper (cellulose, lignin) and adhesives from labels, plastic additives such as pigments and reinforcement materials, and other residues such as food or detergents. Some notable variations between replicates were observed for this parameter, therefore the standard deviation was also reported.

PAG apparent density measurements showed very good reproducibility between replicates (standard deviation < 0.001). The use of optically sorted polymers generated a density gradient, which stretched between 0.919-1.335 g/cm^3. Plastic streams 1-5, which technically consisted of the same materials, have different apparent densities for two reasons. First, a preliminary analysis of all ejected PS and PVC fractions showed that slight variations in apparent density may exist between samples of the same polymer type. Secondly, the ratio of PS to PVC varied over time depending on the concentration of each polymer in the original mixed plastic sample.

The bulk density of PAG was used to calculate a shape index (Eq. 1). An aggregate with a higher shape index can be assumed to show a stronger departure from sphericity and resemble flaky or needle-like particles. Two shape indices are reported for the MIX stream: the first was measured on "coarse" PAG. The second value is for "graded" PAG, namely plastic particles that were sieved to obtain a size distribution similar to sand. The higher shape index seems to indicate that smaller plastic particles were less spherical than larger ones. This is hypothetically the result of the 1.7 mm sieving, which may help concentrate small needle-like particles. Again, the use of optically sorted polymers generated a gradient in shape index, from 62.4 (PE) to 73.6 (PS). Different plastic streams were unique in terms of packaging items. A manual verification of sorted samples showed that thicker injection molded items were dominant in PP and PE streams, whereas PVC and PS streams consisted mostly of thin thermoformed items, such as clamshells and food trays. Assuming that the smallest of the three dimensions of PAG corresponds to the wall thickness of the source packaging item, we concluded that shape index may depend directly on the type of packaging found in a specific stream. Alternatively, the granulation process, which consists of a fast chopping action, could have created several PAG morphologies due to differences in shear strength between various polymers. The bulk density parameter depends on both the apparent density and the shape index. An increase in apparent density will also result in an increase in bulk density, while a higher shape index results in more interstitial space between the particles, thus lowering bulk density. As such, it is a measure of two parameters that may influence concrete mechanical properties.

Table 3.5 : Characteristics of the plastic streams used in this study.

Fraction	% Polymer			Apparent density (g/cm³)	Bulk density (g/cm³)	Shape index
MIX	97.76	±	0.31	0.972	0.334	66.0
					*0.226	*77.0
PP	95.85	±	0.20	0.919	0.330	64.1
PE	97.11	±	0.37	0.952	0.358	62.4
PS	91.09	±	1.31	1.086	0.287	73.6
PVC	97.44	±	0.13	1.335	0.433	67.6
PS-PVC 1	93.79	±	0.53	1.182	0.380	67.9
PS-PVC 2	93.39	±	0.25	1.208	0.395	67.3
PS-PVC 3	94.02	±	0.20	1.200	0.377	68.6
PS-PVC 4	95.07	±	0.11	1.197	0.366	69.5
PS-PVC 5	94.34	±	0.44	1.195	0.349	70.8

*Graded size distribution

3.3.2 Fresh concrete properties

Fresh properties are reported in Table 3.6. Overall, the substitution of spherical sand grains with flakier PAG reduced slump, in agreement with previous work [52, 56]. In Series *a*, which was formulated without using an air-reducing agent, the slump did not decrease as sharply as in Series *b* and *c* for equivalent PAG volumes. The inclusion of PAG caused a substantial amount of entrained air, rather proportionally to the PAG volume. The high air content helped offset the slump reducing effect of PAG by lubricating the mix.

The hydrophobic nature of polymers causes air bubbles to become trapped on the surface of PAG, as these are only partially wetted during concrete preparation. A thin layer of air surrounding PAG could explain the PAG-cement interface gap reported by several authors [58], [59]. The way air bubbles migrate in the cement paste or remain attached to the surface of PAG is not known. However, the strong effect of the air content on concrete compaction, and, in turn, on the compressive strength is well recognized [36].

An air-reducing agent was used in Series *b* mixtures, and the air content was stabilized between 2.5-3.5%, which is comparable to the reference mix (3.7%). The slump values, which were considerably lower than the reference, varied from 40 mm to 10 mm.

For Series *c*, an air-reducing agent was also used to keep entrained air in the same range as Series *b*. Significant variations, however, were observed (2.7-7.6% air content). It is possible that contamination at the surface of the flakes (for example: leftover soap or detergent) generated air bubbles unexpectedly, especially in the case of concrete c5. Plastic samples in this study did not go through a cleaning process as it is not common practice in material recovery facilities.

Table 3.6 : Air content and slump of concrete mixtures.

Concrete mixtures	PAG volume (%)	Air content (%)	Slump (mm)
Ref	0	3.7	125
a05	5	7.2	110
a10	10	8.0	75
a10_graded	10	7.0	35
a20	20	11.2	80
bPP	20	2.8	40
bPE	20	3.1	40
bPVC	20	3.0	25
bPS	20	3.5	10
bMIX	20	2.5	35
c1	10	5.8	60
c2	10	2.7	55
c3	10	3.1	50
c4	10	3.8	50
c5	10	7.6	85

3.3.3 Mechanical properties

3.3.3.1 E-modulus and compressive strength

For series *a* mixtures, the E-modulus and the compressive strength are inversely proportional to PAG volume in the mix (Figure 3.4a). f_c and E fell by 46.9% and 32.1% respectively at 20% PAG volume, compared to the reference mix. The lower strength of *a* series mixtures could be caused at least in part by the very high air content. PAG size distribution did not seem to significantly alter f_c and E (see a10 and a10_graded).

All of series *b* mixtures performed better than concrete a20 for an identical volume of PAG (20% volume of fine aggregates). The use of an air-reducing agent allowed for a significant improvement in f_c and E, although drops in f_c of 13-38% were recorded, respective to the reference mix. PAG have E that are at least an order of magnitude lower than mineral aggregates. Also, since PAG offer very low compressive resistance, they could act as zones of stress concentration in a manner similar to voids in the cement paste, thus helping damage propagation and reducing f_c. Importantly, Series *b* showcased that different plastic streams can significantly influence the mechanical performances of concrete (Figure 3.4b). Ranges of 10 MPa in f_c and 4 GPa in E were obtained for mixtures that are of identical proportioning. Polymers have varying mechanical properties, for example, PVC and PS have a higher E than PP and PE. Interestingly, PVC and PS aggregates respectively gave the higher and the lower limits of spectrum of performances in Series *b*. The results are further investigated with key polymer properties in section 3.4.

Mixtures in Series *c* showed on average better mechanical performances than Series *a* or *b*, since the volume of PAG was relatively low (10% of total fine aggregate volume) and an air-reducing agent was used (Figure 3.4c). E was 12-18% lower than the reference mix. Series *c* showcased a good repeatability in E with PAG collected from the same material streams over a period of 9 months. Values ranged between 27.68 GPa (c5) and 30.08 GPa (c3). More variation was observed in f_c, where values ranged from 31.81 MPa (c5) to 40.85 MPa (c3). Concrete c5 had a very high air content (7.6%), compared to an average of 3.9% for others in Series *c*, which probably resulted in a poorer structure. This suggests that uncertainties arise from the use of unwashed contaminated plastic.

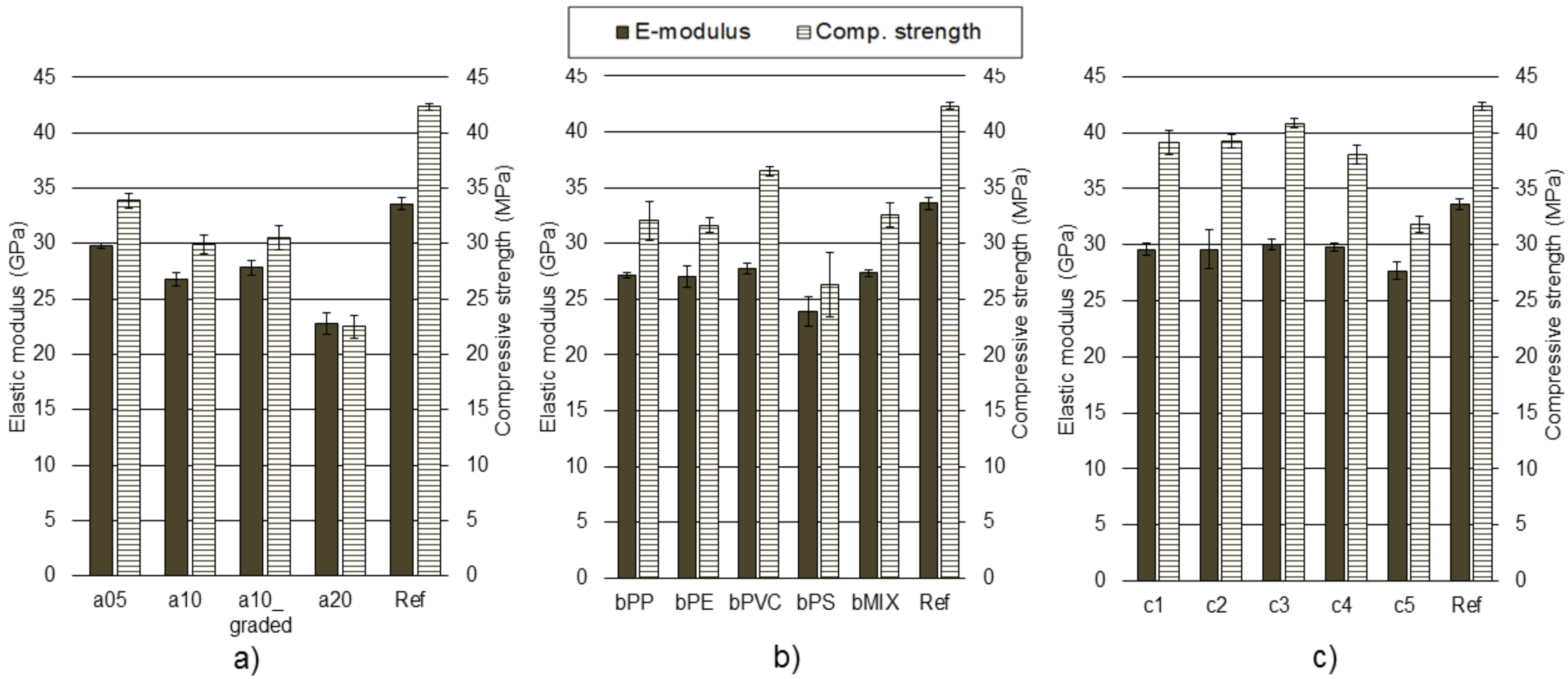

Figure 3.4 : E and f_c for series a, series b and series c. Standard deviation bars shown (5 replicates).

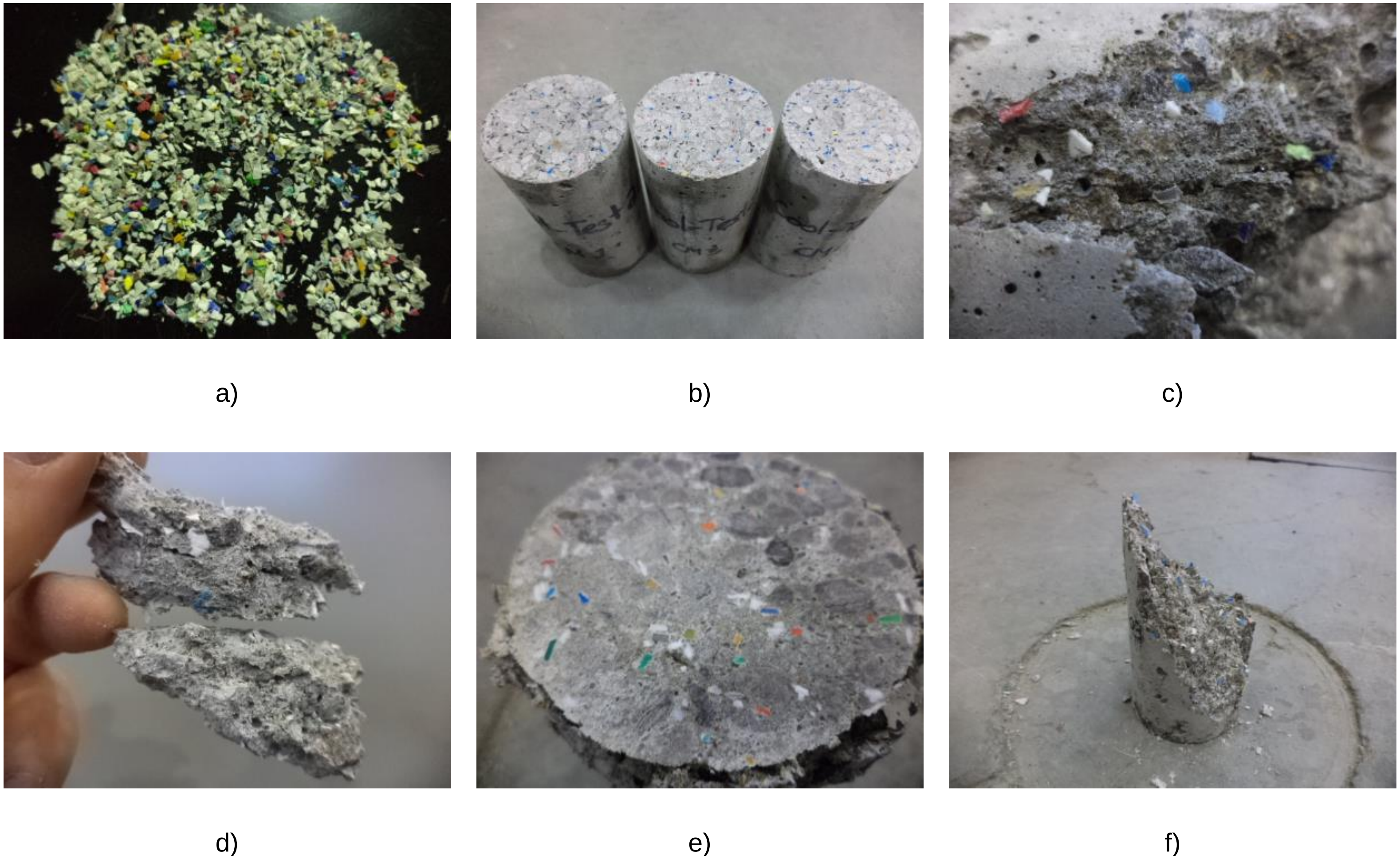

Figure 3.5 : a) PP aggregates; b) Cylinders before mechanical testing; c)-f) Cylinder fragments after mechanical testing.

3.3.3.2 Post-cracking strength (PCS)

Figures 3.6a)-3.6d) show the mean compression stress-strain curves used to evaluate PCS. Series *a* averaged stress-strain curves seem to follow a trajectory similar to the reference mix curve (Figure 3.6a). A look at 90% confidence interval bands (Figure 3.6b, n=5), however, shows a much narrower data dispersion after the resistance peak in PAG mixtures (the same phenomenon was observed in Series *b* and *c*). The confidence interval for the reference mix (n = 10) quickly expands after peak resistance because brittle failure occurred randomly in the replicates. It is possible that PAG are similar to polymeric reinforcement fibers in helping to dissipate fracture energy and acting as bridges across cracks to slow down damage propagation [54]. Figure 3.5d) seems to illustrate an example of this bridging action. It is also noteworthy that graded PAG were more efficient than coarse PAG in maintaining PCS. Assuming that residual strength and ductility in PAG mixtures is attributed to plastic flakes delaying crack coalescence and propagation, smaller flakes seem to further reduce crack propagation by generating more reinforcing particles for the same plastic volume.

Overall, significant performance gains were observed in Series *b* and *c* because of lower entrained air. Moreover, PS and PVC aggregates in Series *b* (and their combined use in Series *c*) generated mixtures that exhibit a higher strain at f_c than the reference mix. As a result, at 10% PAG volume, experimental mixtures of Series *c* had W_e that were equal or superior to the reference mix, except for the high entrained-air c5 mixture (Table 3.7).

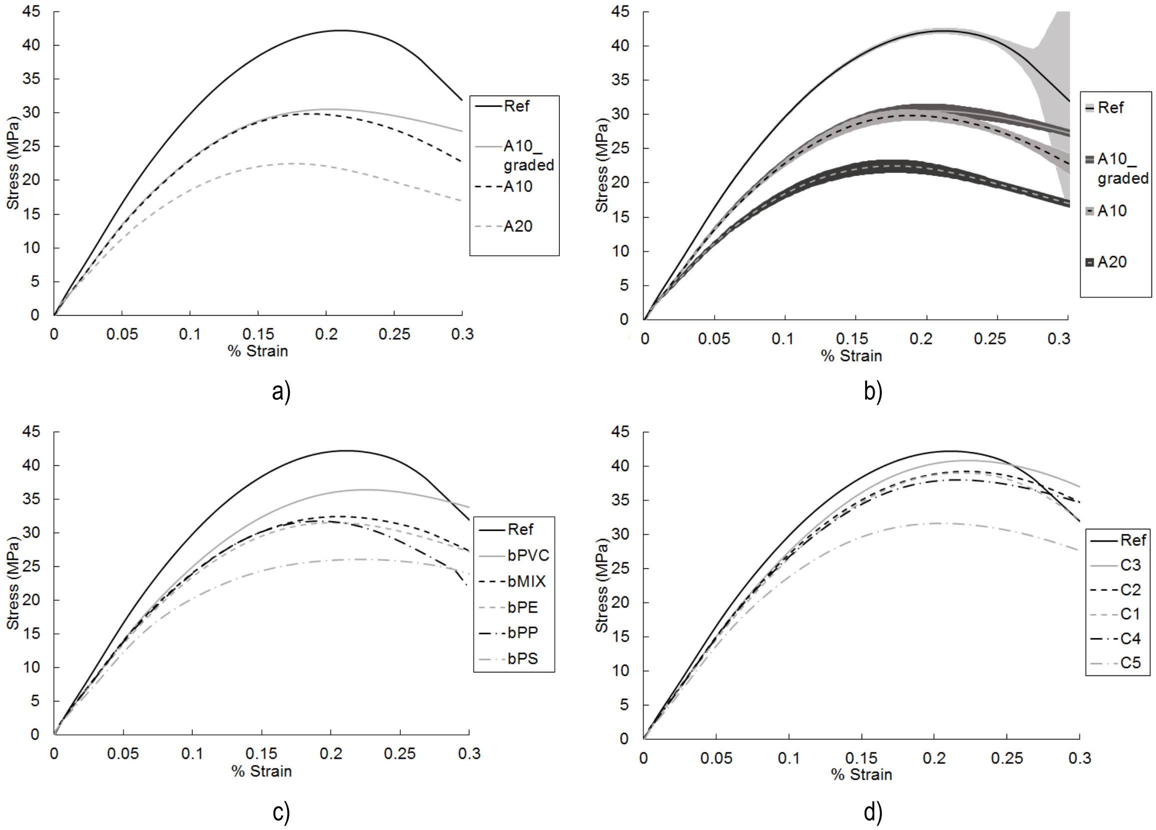

Figure 3.6 : Mean compressive stress-strain curve for (a) series a; (b) series a with confidence interval bands (90%); (c) series b; (d) series c.

Table 3.7 : Toughness indices for reference and experimental concrete mixtures.

Concrete mix	W_e ($J \cdot m^{-3}$)	I_3	I_5
Ref	0.64	7.22	14.39
a10	0.44	7.11	14.34
a10_graded	0.39	7.26	15.37
a20	0.19	7.06	15.63
bMIX	0.50	6.93	14.14
bPE	0.48	7.03	14.34
bPP	0.51	6.98	13.70
bPS	0.45	6.57	13.23
bPVC	0.62	7.10	14.51
c1	0.63	7.29	14.70
c2	0.72	7.07	13.78
c3	0.74	7.20	13.84
c4	0.65	7.16	14.43
c5	0.48	7.10	14.50

3.3.3.3 Dry density, water absorption and thermal conductivity (λ)

Six mix designs were chosen for further analyses of dry density, % water absorption, and λ: Ref, a10, a10_graded, a20, bMIX and c2. These were selected with the aim of studying water absorption and λ parameters over a large gradient of concrete dry density.

Dry density values progressively decreased with higher air and plastic content, and ranged between 2.26 g/cm^3 for the reference mix (3.4% air, 0% PAG) and 2.00 g/cm^3 for a20 (11.2% air, 20% PAG). Variations in water absorption by immersion, however, showed unexpected results. Indeed, mixtures with a very high air content such as Series *a* should normally be more porous and permeable, leading to more water absorption. Despite a lack of data on air content in concrete containing PAG [35], previous work has shown that adding PAG generate voids. Marzouk *et* al. used scanning electron microscopy to reveal that interfacial zones are larger between the cement matrix and PAG compared to sand [58]. This observation was corroborated in the same study by higher gas permeability and water absorption results for mixtures with high PAG contents. Hannawi *et* al. [57] observed similar interfacial zone characteristics, but reported lower coefficients of water sorptivity with higher PAG contents. The authors attributed these results to the hydrophobic nature of the polymer flakes, which helped slow down the water imbibition front. Our results (Figure 3.7a) seem to agree with the latter study, as the amount of absorbed water was highest in the reference mix (8%), and lowest in a20 (3.5%). The difference in water absorption between the almost identical a20 and bMIX could be attributed to the use of an air-reducing surfactant in the latter, which may have had the effect of chemically altering PAG surfaces, thus reducing their hydrophobicity.

These results suggested that investigating voids content and porosity by water immersion methods may not be suitable for mixtures containing PAG, since a complete saturation might not be possible if air bubbles remain attached to the polymer flakes. Silva et al. (2013) also reported inconsistencies in using this method for experimental mixtures containing PET flakes, and suggested that immersion testing be conducted under vacuum conditions to ensure a full saturation of test specimens [72].

The mean thermal conductivities are given in Figure 3.7b. Similar to our results, positive relationships between λ and density of concrete were also reported in previous studies. Demirboga et al. [66] suggested that λ was mostly influenced by the increased porosity and air voids that PAG help create in concrete, since air has a much lower λ than other constituents, and others [54, 61] attributed their results to a combination of the lower conductivities of both polymers and air voids.

Our results seem to agree with the last hypothesis. Series *a* mixtures, with high air contents, are found at the lower end of the conductivity spectrum. c2 and bMIX, however, have significantly lower thermal conductivity than Ref despite having lower but similar air contents. Thermal conductivity could therefore be well approximated as an average of the conductivities of all concrete constituents, including air and PAG volume, much like concrete dry density. Côté and Konrad (2005) suggested that thermal conductivities in cemented materials are also influenced by the shape of material pores, and the ratio of water to air trapped inside [71]. Providing that differently sized PAG would generate porosities of varying geometries, a more thorough investigation is needed on this topic.

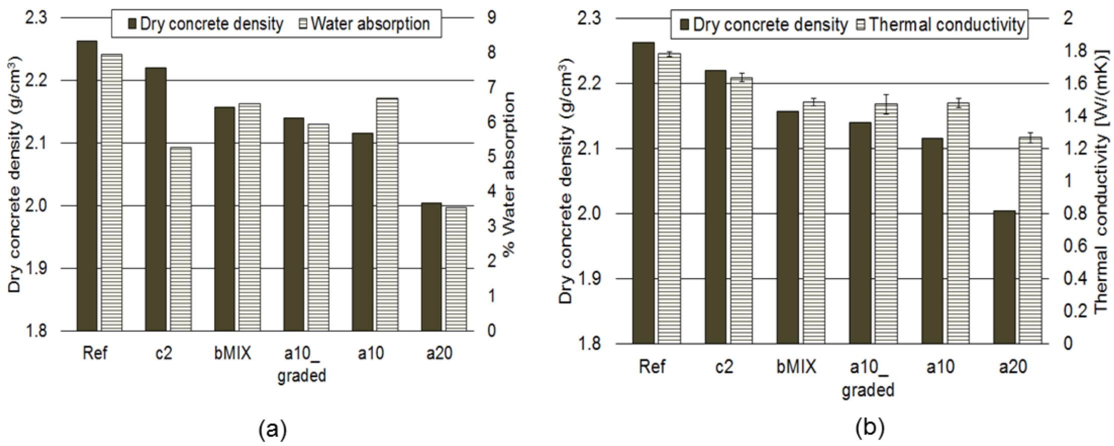

Figure 3.7 : For selected mixtures: (a) Dry density and % absorbed water; (b) Dry density and thermal conductivity.

3.3.4 Correlation study

This section highlights the results of a correlation study between mechanical performances and PAG characteristics for experimental series *a*, *b* and *c*, as well as for the six mix designs selected for the thermal conductivity and absorption investigation (see section 3.3.3). Pearson correlations tables were computed, and the most relevant correlations are plotted and discussed in Figures 3.8 to 3.12 below. Complete correlation tables are found in Annexe 3.

The observed reduction of f_c in Series *a* mixtures is explained in great part by air entrained in the mix by hydrophobic plastic particles, as evidenced by the strong correlation between air content and PAG volume. Given that there is a lack of information on measured air content in previous studies on PAG concrete [35], further work on the topic should take variations of entrained air into account. Additionally, the use of an air-reducing concrete additive proved to be effective in enhancing PAG concrete performances.

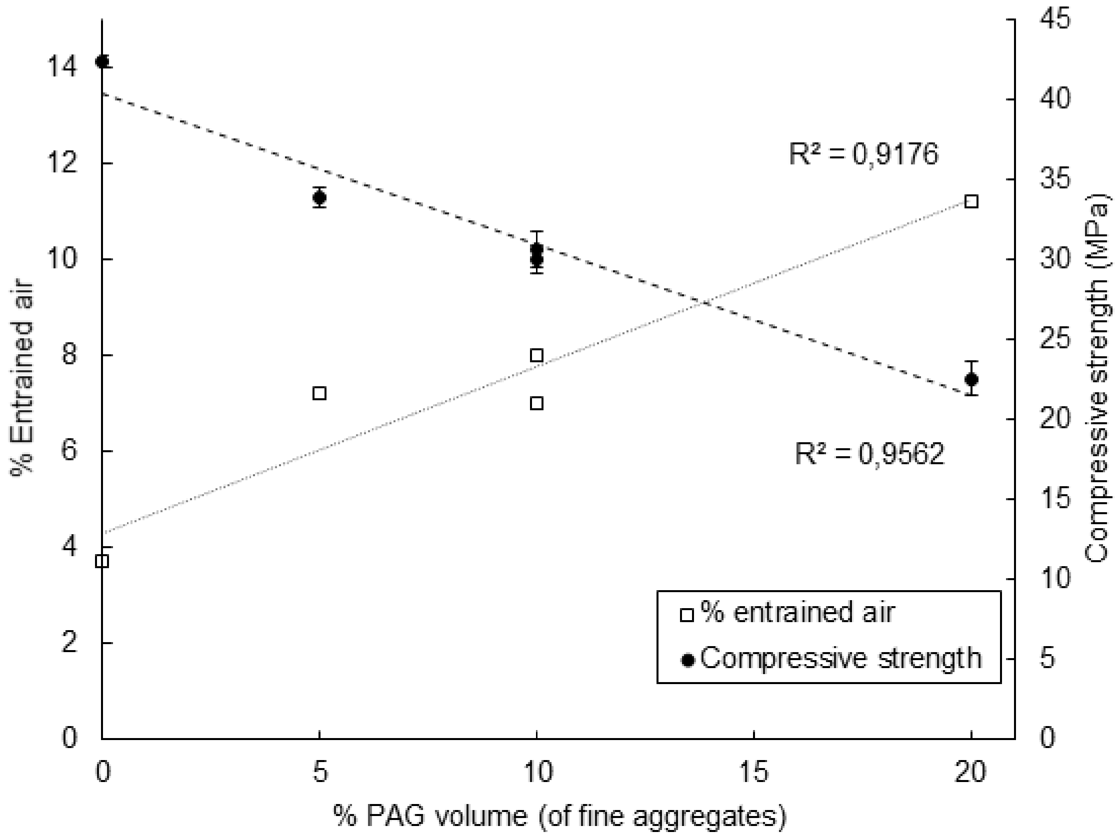

Figure 3.8 : Series a f_c and % air content vs. PAG volume. Standard deviation error bars are shown for f_c (n = 5).

In Series *b*, PAG shape index varied strongly between polymeric fractions since they consist of very different plastic packaging, as discussed in section 3.1. In parallel, it has been demonstrated that slump is partly dependent on the shape (angularity) of aggregates in the mix. PP and PE fractions consisted mostly of thicker injection-molded items such as buckets, pails or food tubs and lids. The PS fraction consisted mostly of thin-walled thermoformed items such as yogurt containers or disposable cups. Although the same granulation process and sieves were used to generate all PAG, these inherent differences in container wall thicknesses generated particles of different morphologies, with the resulting effects on fresh concrete slump (Figure 3.9). In further work, some material stream characteristics such as plastic packaging types could be measured to optimize and achieve consistent concrete workability.

Aggregate bulk density accounts for both aggregate packing (which is dependent on shape index) and the intrinsic apparent density (ρ) of the material. This parameter seems to be strongly related to f_c for Series *b* (Figure 3.10a) and, although to a lesser extent, Series *c* (Figure 3.10b). Neville elaborated on the importance of aggregate characteristics such as shape, E and surface porosity in concrete mechanical properties [36]. Mainly, spherical aggregates of a wide size distribution lead to mixtures with closely packed aggregates and better mechanical resistance, while the same aggregate density is not possible for randomly distributed elongated or flaky particles. Aggregate shape alone may explain up to 22% of concrete f_c and 31% of concrete f_t [36]. Material density on the other hand, is positively related to mechanical properties such as E [73]. Taken together, aggregate mechanical resistance and shape are very important drivers of concrete mechanical properties, making bulk density a particularly important aggregate characteristic to be considered.

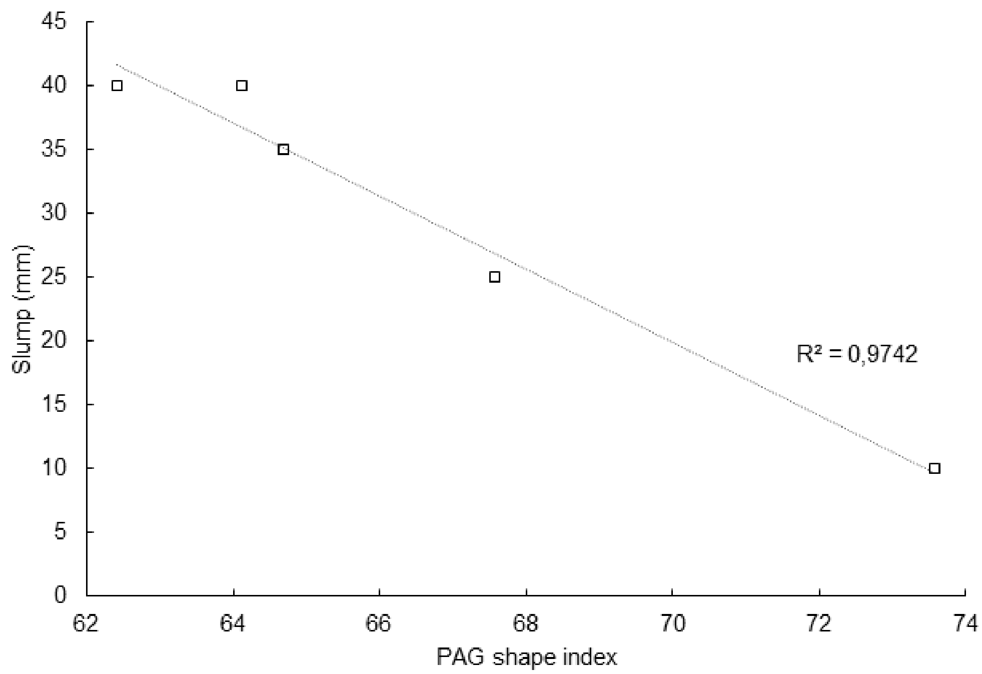

Figure 3.9 : Slump vs. PAG shape index for Series b.

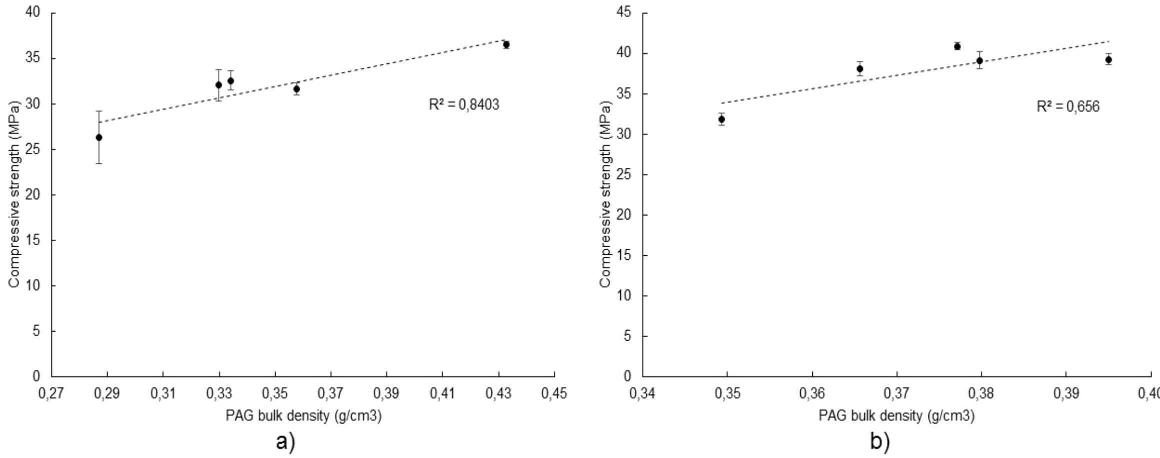

Figure 3.10 : a) fc vs. PAG bulk density for Series b. Standard deviation bars (n = 5) shown. b) f_c vs. PAG bulk density for Series c. Standard deviation bars (n = 4) are shown.

Entrained air was strongly correlated to mechanical properties in Series *c* (Figure 3.11). Like Series *a*, there is a large range of entrained air between mixtures, despite the use of an air-reducing agent at low concentrations. The percentage of entrained air does not seem to be strongly dependent on any of the measured PAG properties (shape, % polymer, apparent and bulk densities). Surface contamination of PAG from material stream contaminants such as residues in household detergent of soap bottles remains the most likely explanation for varying air content. It may be advisable to include a washing step in the preparation of PAG to achieve consistent results in further research. The positive relationship between slump and % air content is also evidenced in Figure 3.11, but the effect of the former on concrete resistance is not clear and likely not significant.

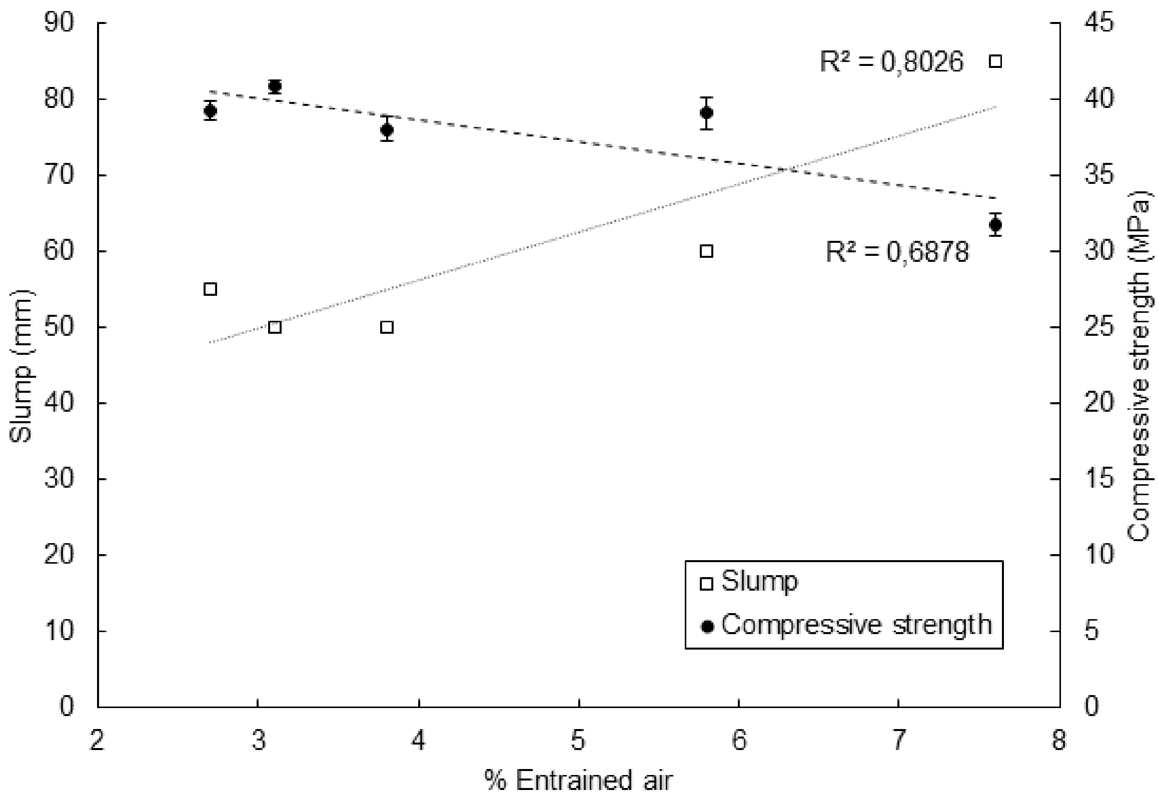

Figure 3.11 : Slump and f_c vs. % entrained air in c series.

The relationship between concrete dry density and λ is the only strong correlation obtained for the six selected mixtures (section 3.3.3). Changes in dry density resulted from a combination of varying PAG volumes and % entrained air. Air has a low λ (0.024 W/ (mK)) and air voids would therefore help decrease heat flux. PAG has a similar effect since polymer thermal conductivities are about one order of magnitude lower than conventional concrete materials. Given that a high air content has a deleterious effect on mechanical properties, whereas PAG generate possible benefits for concrete toughness and PCS, a concrete with a low air content but incorporating some plastic could help in reaching an interesting compromise between mechanical and thermal properties.

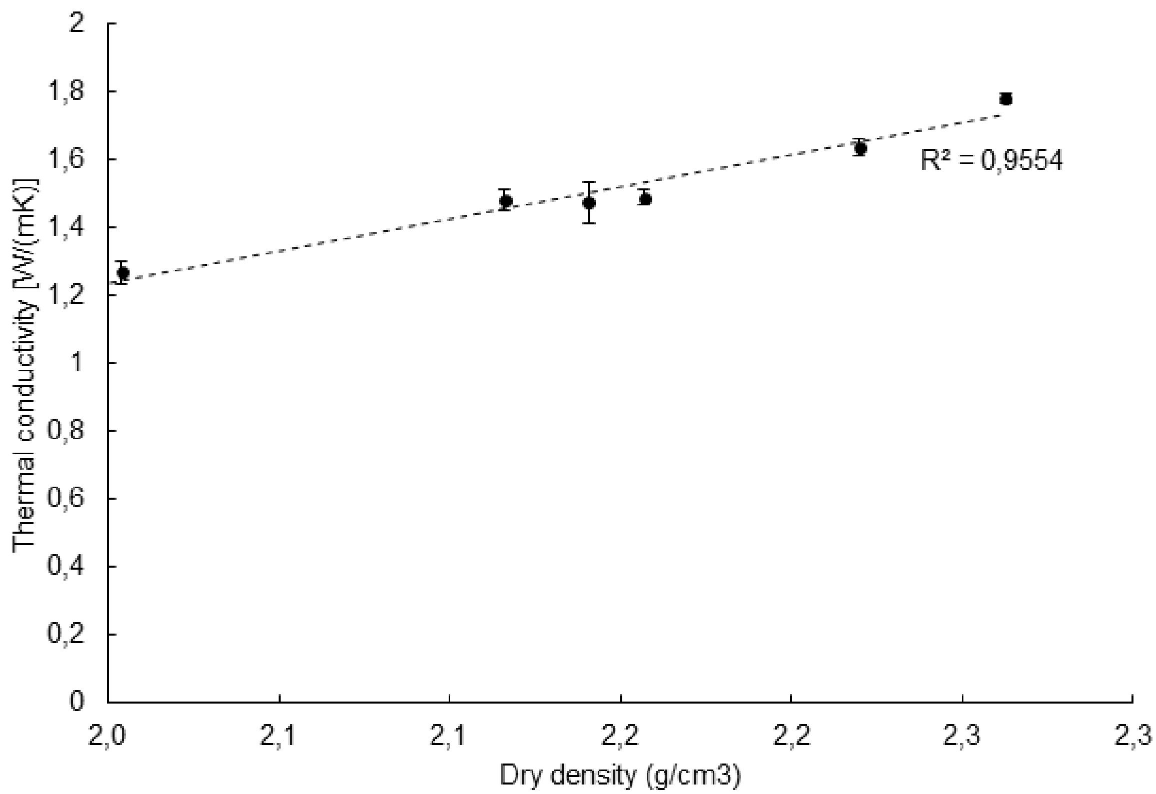

Figure 3.12 : Concrete dry density vs. λ. Standard deviation bars are shown (n = 2).

3.4 Conclusion

This work analysed the properties of concrete incorporating PAG from representative samples of a postconsumer material stream. The effects of the various mixture designs with varying PAG volume, as well as the effects of PAG size, polymer type and other characteristics on concrete were studied. Material stream variations were also considered using a series of plastic samples collected over one year.

Mechanical performances (E and f_c) were negatively impacted by increasing PAG volumes. Concurrently, PAG have the effect of entraining substantial air, which accounts for part of the observed performance drops. The double effect of entrained air and PAG, however, generated mixtures with a lower λ when compared to the reference mix. Particle size was found to have the strongest observable effects when studying the post-cracking resistance of concrete, but almost no influence on concrete E, f_c, λ or water absorption.

PAG generated from different polymer fractions had different characteristics and it impacted concrete performance. PVC aggregates were found to perform the best, whereas PS aggregates had poor performances in terms of E, f_c and W_e. A correlation study between selected PAG properties (apparent and bulk densities, % polymer content, shape index) and concrete characteristics helped reveal that PAG bulk density is an important underlying factor for disparities in mechanical performances, while shape index had a strong influence on concrete workability (slump).

Sampling material over time generated some disparities in material composition and PAG characteristics. Unexpected variations in entrained air, likely due to PAG surface contamination, modified concrete f_c and slump and may have occulted other correlations between PAG characteristics and concrete performance.

Incorporating plastic in concrete is a promising recycling strategy since it eliminates the expensive requirement of sorting plastics by polymer type used in conventional closed-loop recycling. Indeed, mixed polymer aggregates generated mixtures that are relatively similar to the ones made with a single polymer type.

Material stream characteristics of importance in designing mixtures are PAG shape, bulk density and volume in the mix. Uncertainties can be reduced by a careful monitoring of variations in plastic packaging types in the material stream. Furthermore, it may be necessary to include a washing step if the stream is contaminated by chemicals that may react with concrete ingredients such as water. Lastly, the use of an air-reducing agent will help stabilise air content and enhance mechanical performances considerably.