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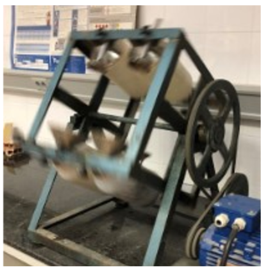
TECHNICAL AND ENVIRONMENTAL FEASIBILITY OF RECYCLED PLASTIC FIBRES FROM FOOD PACKAGING WASTES AS REINFORCEMENT IN CONCRETE

PIETRO ANTONIO VACCARO

Sevilla, 2022



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Memoria de tesis presentada por Pietro Antonio Vaccaro
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A mi familia

To my family

"Fatti non foste a viver come bruti ma per seguir virtute e canoscenza"

— Dante Aleghieri.

"Longum iter est per praecepta, breve et efficax per exempla"

— Lucio Anneo Séneca.

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ABSTRACT

The growing and current concern for the enormous quantities of plastic waste produced each year, and the now inevitable daily use of plastic, has attracted the attention of more and more researchers for many years. Plastic waste can follow different paths in order to be disposed or be recycled or transformed into energy. The plastic packaging sector occupies the first place in Europe with regard to the use of plastic, and for this reason, most of the studied have focused on it in order to find solutions at the end of life of this material. Within plastic packaging, a large part of the market is made up of food plastic packaging. Its countless advantages have been developed in recent years thanks to new technologies and new material products. Among these, it can be found that the films used in food plastic packaging can be monolayer, made with a single material, or multilayer, which instead consist of two or more layers of different materials for each layer. The latter offer numerous advantages over the former because multiple functions and requirements are met at the same time. Simultaneously, however, numerous problems appears when they become waste difficult for managing because their recycling is difficult, due to it is made up of materials with different characteristics.

The main objective of this doctoral thesis is to help find a new end-of-life to multilayer food plastic packaging waste and, at the same time to deal with the use of innovative materials in the engineering and construction sector.

This research was addressed on two points: from a technical nature, aiming at ascertaining whether the new material tested, that is the concrete reinforced with recycled plastic fibres (RPF) obtained from multilayer films of industrial waste from food plastic packaging, had the same physical characteristics and mechanical performance compared to ordinary concrete; and from an environmental nature, aiming at assessing the impact on environment that this new material could have based on leaching tests.

The research was divided into the following phases:

1. A first theoretical phase of bibliographic and regulatory investigation on the subject of the doctoral thesis, during which a bibliographic compilation and revision of the updated literature on the topic was carried out;
2. A second phase, which was experimental, from a feasibility and technical perspective, that developed through the acquisition of plastic waste from a Spanish company operating in the packaging sector, with which the macro plastic fibres used were obtained and prepared, and then added to the mixture for the production of the concrete specimens necessary to be subjected to physical and mechanical tests. The specimens produced were cubic, cylindrical, and prismatic with a fibre content of 2 kg/m^3, 4 kg/m^3 and 6 kg/m^3. Two types of control specimens were also made: a control specimen of concrete without reinforcement, and fibre-reinforced concrete made with a commercial polypropylene fibre with a dosage of 2 kg per m^3 of concrete. From the results of the compression and bending tests, the respective resistance values at 7 and 28 days, the modulus of elasticity and the toughness of the material have been obtained. Physical-chemical analyses were also carried out on concrete specimens.
3. And an also final experimental phase were different types of leaching tests were performed: (1) basic characterization on RPF, and (2) maximum availability and (3) leaching test for the

manufactured concrete in monolithic state.

Following the processing, analysis and comparison of the results obtained from the tests, the following conclusions could be drawn:

1) mechanical:

- The physical properties studied were not significantly affected by the addition of recycled plastic fibres;
- The addition of RPF produced a certain decrease in compressive and flexural strength, but an improvement in the post-cracking properties of the concrete was achieved;
- The increase in toughness index with an increase in the percentage of recycled plastic fibres confirmed their stitching action inside the cement matrix;
- The toughness index presented by the concrete with commercial fibres was greater than that with the same dosage of RPF, due to its rougher surface and greater adherence to the cementitious matrix. Furthermore, it should be noted that the toughness index relative to the mixture with 6 kg of recycled plastic fibres per cubic meter of concrete was comparable to that with commercial plastic fibre (PFRC-REF). This suggests that manufacturing RPF with a rougher surface could lead to a reduction in the amount needed to achieve similar toughness index levels to PFRC-REF;

2) environmental:

- Waste plastic food packaging, used for the preparation of plastic sheets to be used as fibre reinforcement in concrete, showed low release levels of the elements determined by the Council Decision 2003/33/EC and they were under the inert limits, except for antimony. Hence, they were classified as non-hazardous material;
- The low release levels in concrete with RPF confirms that its incorporation into concrete reduced the concentrations in leachates to a minimum. The concentrations of the elements with higher mobility, in the long-term, were much lower than the limits imposed by regulations for "shaped" construction materials, such as Soil Quality Degree (from The Netherlands);
- The study carried out to identify the transport and chemical mechanisms, indicated that the release patterns were: wash-off for Sb, Zn (although Zn also presented depletion in the final steps) and Cr in concrete samples with RPF; dissolution for Ba, except for FRC-REF samples; and diffusion for Cr in FRC-REF and C-REF samples;
- This type of material did not present any possible environmental impact and could be a viable alternative that would help increase the valorisation of such types of plastic waste, which is considered as being one of the biggest concerns worldwide.

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ABBREVATIONS

CEN/TC	CEN Technical Committee
CEN-TS 15862:2012	European Committee for Standardization - Technical Specification
CMOD	Crack mouth opening displacement
CPF	Commercial polypropylene fibre
C-REF	Concrete without reinforcement
EEA	European Environment Agency
ENAC	Entidad Nacional de Acreditación
EPS	Sintered expanded polystyrene
FPW	Food packaging waste
FRC	Fibre reinforced concrete
FRC2-REF	Fibre reinforced concrete made with commercial fibre dosage of 2 kg/m^3
FRC-REF	Fibre reinforced concrete for reference
FRP	Fibre reinforced plastic
HDPE	High- density polyethylene
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
LDPE	Low-density polyethylene
LLDPE	Linear Low-Density Polyethylene
LVDTs	Linear variable displacement transducers
MDPE	Medium-density polyethylene
MLP	Multilayer plastic
NEN 7371	Dutch standard extraction test
OPA STE	Biaxially oriented polyamide
PA	Polyamide
PAN	Polyacrylonitrile
PC	Polycarbonate
PE	Polyethylene
PE-HD/PE-MD	Polyethylene, high and medium density
PE-LD/PE-LLD	Polyethylene, low and liear density
PET	Polyethylene terephthalate
PF	Plastic fibres
PF	Polypropylene fibres

PFRC	Plastic fibre reinforced concrete
PMMA	Polymethylmethacrylate
PP	Polypropylene
PFRC-REF	Concrete made with commercial polypropylene fibres
PS	Polystyrene
PUR	Polyurethane
PVC	Polyvinyl chloride
PVDC	Polyvinylidene Chloride
RPF	Recycled Plastic Fibres
RPFs	Concrete reinforced with macro plastic fibres
SA	Strategic activities
SDG	Sustainable Development Goals
SQD	Soil Quality Decree
ULDPE	Ultra Low-Density Polyethylene

1. Introduction

1.1 Background

1.1.1 A brief history of plastic

The origin of plastic, understood as a natural polymer, dates back to ancient times, when Human began to use natural products such as amber, tortoise shell, horn [1], bones for his own purposes of animals, silk, wool [2], plant cellulose or starches [3].

The first semi-synthetic plastic material, Parkesine (also called synthetic ivory) was patented by the English chemist Alexander Parkes in the second half of the 19th century, following the development of studies on cellulose nitrate [3]. In Figure 1.1(a) we see the outside of the factory where Alexander Parkes produced the first plastic in the world [4] while Figure 1.1 (b) shows the Bronze medal that was given to the English chemist on the occasion of the International Exhibition in London in 1862 [5].

(a)

(b)

Figure 1.1 The Parkesine factory at Hackney Wick, London (a) [4] and the replica of the Bronze medal awarded to Alexander Parkes at the International Exhibition in 1862 in London (b) [5].

In 1869, the English Daniel Spill, former partner of Alexander Parkes, founded a company for the production of Xylonite, a plastic material that took the place of Parkesine [1]. Subsequently, in 1870, the American Hyatt brothers patented the celluloid formula [3,6], the aim was to replace the expensive and increasingly rare ivory used to build billiard balls with a cheaper material [1]. Celluloid will later be used in many other sectors: production of cinematographic films, material for use by dentists for making dental impressions [6], waterproofing of the wings and fuselages of the first aircrafts [7].

Nevertheless, it is from the early years of the twentieth century that plastic obtains its greatest successes. Starting from 1909, with the Bakelite patent by the Belgian chemist Leo Baekeland, indispensable objects for the nascent automotive industry were produced such as electrical parts [2,6,12], brake pads and pistons [8], as well as to everyday objects such as microwave pans, handles, bottle caps, linings [10,11,12]. The success of bakelite was essentially due to its remarkable resistance to electricity, heat and chemical action [12]. In Figure 1.2, it can be observed some objects made with a Bakelite casing: in Figure 1.2 (a) a Siemens telephone from 1929 [9] is depicted, in Figure 1.2 (b) a pocket camera produced in the mid-1930s [10] while in Figure 1.2 (c) is shown one radio with valves of the 1950s [11].

(a) **(b)** **(c)**

Figure 1.2 Objects made with Bakelite: Siemens telephone (a) [9]; pocket camera (b) [10]; radio with valves (c) [11].

In the following years, many other synthetic plastics were produced: Polyvinyl Chloride (PVC), in 1912, by the German chemist Fritz Klatte [13]; Cellophane, in 1913, invented by the Swiss Jacques Edwin Brandenberger. Additionally, it was precisely when, from this last material produced in very thin and flexible transparent sheets, the development of packaging began [14,15].

At the beginning of the 1930s, with the emergence of the new raw material, oil, the development of synthetic materials derived from the petrochemical industry began. In 1935, the American chemist Wallace Carothers synthesized Nylon, a new synthetic fibre, that will be used mainly by the American and British textile industry in order to make up for the import of silk and for the production of parachutes, women's socks and ropes [1,16]. British chemists Rex Whinfield and James Tennant Dickson, in 1941, patented the first polyester fibre, Polyethylene Terephthalate (PET) [17], a material that, both for toughness and resilience, was superior to nylon [18]. Its use proved to be very useful in the military field, as a substitute for metals and natural materials [19]. In the early 1950s, the Italian chemical engineer Giulio Natta, together with the German chemist Karl Ziegler, developed a thermoplastic polymer, Isotactic Polypropylene (PP), which due to its dimensional stability, resistance to chemical products and a considerable duration at high temperatures, it will find various applications [20]. In Figure 1.3 you can see some examples of objects currently made with PP: weldable polypropylene fittings, designed for the construction of hot and cold sanitary water distribution systems [21], other everyday objects [22,23].

(a) **(b)** **(c)**

Figure 1.3 Polypropylene objects: fitting for plumbing systems (a) [21]; objects of wide daily use (b) [22]; seats of a stadium (c) [23].

In 1973, the American engineer Nathaniel Wyeth created a variant of PET capable of withstanding the pressure of carbonated liquids: thus, the era of food packaging and the PET bottle, light, impact resistant and transparent, was born [24]. Figure 1.4 shows the variety of shapes, sizes, colours and uses of the containers that can be made using PET [25,26].

(a) **(b)**

Figure 1.4 Containers in Polyethylene Terephthalate: plastic bottles for liquid packaging (a) [25]; products specially designed for pharmacies and healthcare professionals (b) [26].

Starting from the 1960s and up to modern times, thanks to the development of new technologies, technopolymers (called engineering polymers) established themselves [27]. Figure 1.5 shows the relative importance of the materials used in engineering over time, divided by type (metals, polymers and elastomers, composites, ceramics, glass). In this diagram, the abscissa axis is time, in a non-linear scale, and on the ordinate axis is the relative importance of the materials, a magnitude assessed on the basis of the historical period: for the period that includes the Stone and Bronze Ages its value was established by archaeologists; in 1960 is based on the teaching hours of the study of materials carried out at the universities of the United Kingdom and the United States; in 2020 on the forecasts of the use of materials in cars by manufacturers. It is highlighted that starting from prehistory and up to the end of the 1950s, there has been a continuous decline in their relative importance and that instead, after the Second World War, following the post-war industrial recovery, the trend has reversed [88].

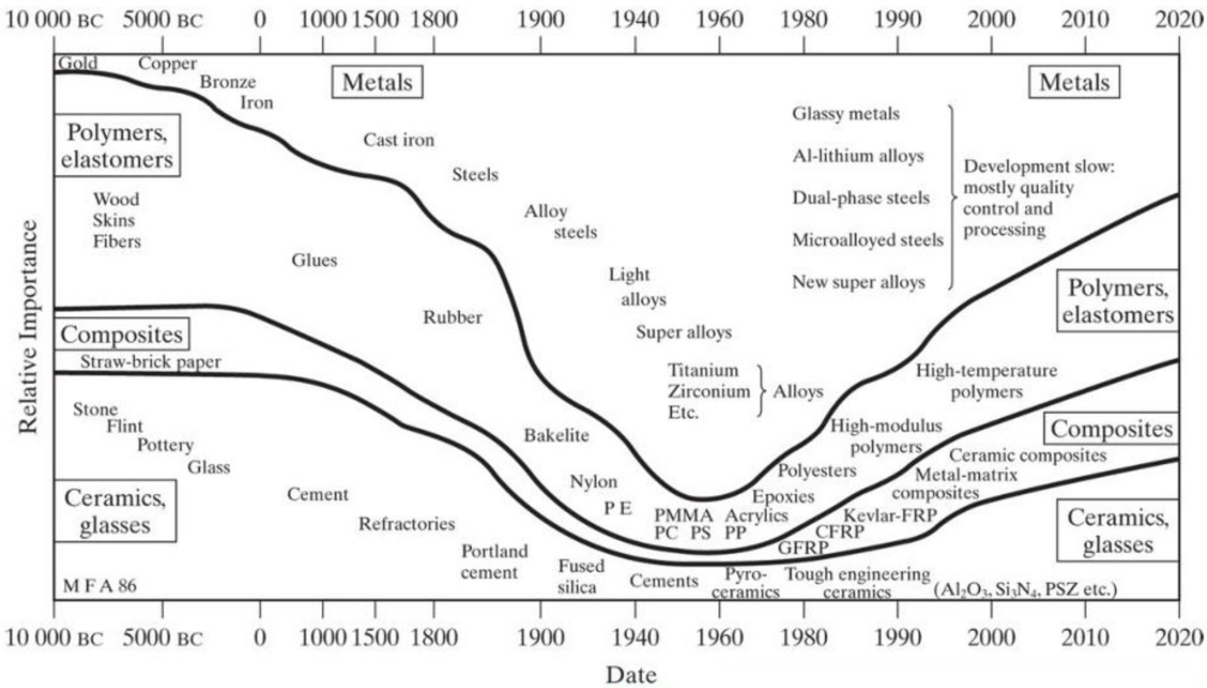


Figure 1.5 The change in the relative importance of materials used in engineering over time [88].

These plastics, due to their high physical-mechanical characteristics (stiffness, toughness, ductility, workability, resistance to high temperatures, static and dynamic loads or aging) allow to be used in place of metals [27], in many cases. They are used in almost all fields of human activities, from the manufacture of items for clinical laboratories [28], to the automotive industry for engine components [29], to the production of space helmets for astronauts [30], to the production of turbine blades and other components of the engines of jet aircraft [31, 32], contact lenses [33, 34], in the production of pistons and piston rings for automobiles [35, 36], mobility [37, 38], ecology industries [39] and in the food industry [40].

Subsequently, it is precisely the problem of residues in the food packaging industry, where plastics in the form of monolayer and multilayer films are widely used, that this research aims to address and make its contribution. In Figure 1.6 it can be seen some examples of components and objects made with technopolymers which, thanks to its high features, allows them to be used for a wide variety of purposes.

(a) **(b)** **(c)** **(d)** **(e)**

Figure 1.6 Some objects made with technopolymers: plastic engine (a) [36]; gears for food machineries (b) [40]; drone (c) [37]; New Jersey Road Separators (d) [38]; waste container lids (e) [39].

1.1.2 Classification and application of plastics

Since the vast variety of polymers, each with different properties, characteristics and fields of use, "plastics" have been the subject of worldwide legislation. In Spain, the UNE EN ISO 1043-1:2012/A1:2016 [41] establishes that a code is associated with each plastic material, which uniquely identifies it. Regarding the classification of plastics, if it is considered the nature of the raw materials from which the polymer is obtained, it can be can distinguished polymers into: natural, synthetic and partially synthetic [57]. The natural ones are those that occur in nature or are produced by living organisms, such as cellulose, silk, chitin, protein or DNA [58]. Partially synthetic polymers are obtained by chemically modifying natural polymers while synthetic ones are obtained from petroleum refining products by chemical polymerization processes [57,59]. On the other hand, if it is considered the behaviour of the plastic material towards heat, it can be distinguished the polymers in thermoplastic and thermosetting.

Thermoplastics are those which, when heated, soften until they liquefy but, if cooled, harden again and, therefore, can be subjected to repeated heating and subsequent cooling cycles without losing most of the initial features. Thermosetting polymers, once permanently hardened in the forming process, if reheated they become insoluble and infusible [57].

The distribution and fields of use of polymers produced in Europe is shown in Figure 1.7. It can be seen how the most widespread plastic material in Europe is the PE. Both the PE-LD/PE-LLD and the PE-HD/PE-MD make up 29.8 % of the European consumption of plastics. In that case, the PP is the second plastic material used. PE, PP and PVC represent 59.2% of the total plastic used in the European market in the year 2019 [45].

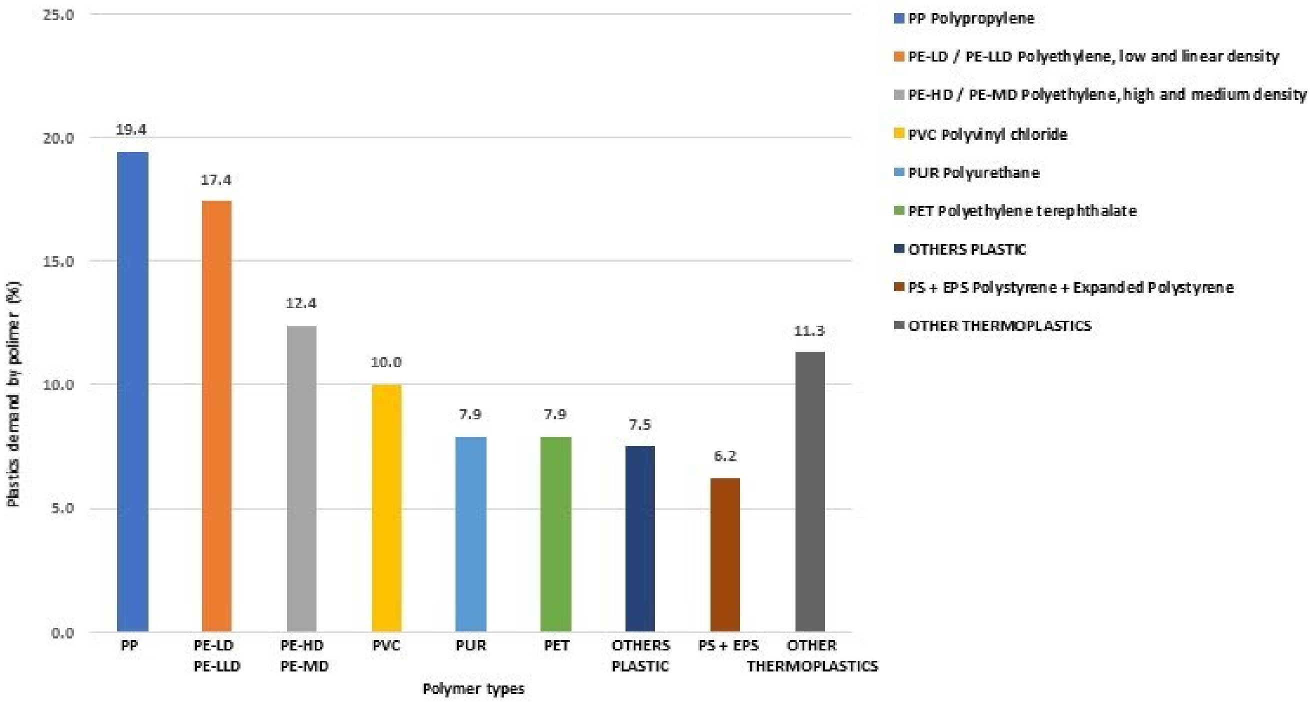


Figure 1.7 The distribution and fields of use of plastics produced in Europe [45].

Figure 1.8 shows the sectors in which plastic is used and the relative percentage of use compared to the 50.7 million tons of plastic of European demand in 2019. Spanish demand, in the same year, is 7.8% of the European one [45]. The data shows how the plastic market is largely oriented towards packaging followed immediately after by the building and construction sector.

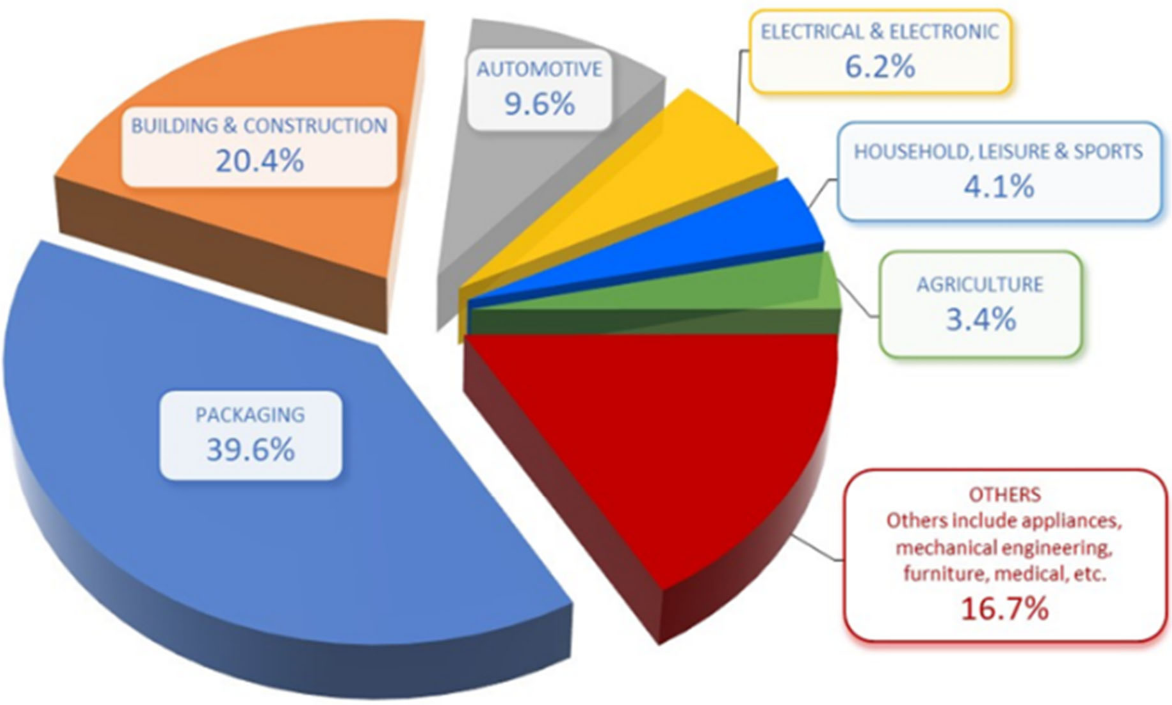


Figure 1.8 The demand for plastics in Europe, divided by sectors [45].

Within the plastic packaging sector and, specifically, in food packaging, multilayer films [69,70] were created starting from the 90's, flexible and rigid packaging, consisting of a package of two or more layers of different thicknesses and materials, glued together, which in addition to plastic use materials such as aluminium or cardboard. And it is precisely the recycling of industrial waste from this type of multilayer food packaging that is involved in the present research.

Table 1.1 shows the abbreviations of the main plastic materials currently most in use in the field of civil constructions (construction, transport or plumbing) and, particularly, in those which, in the form of fibres in addition to the traditional components of concrete, find application in the construction of structural and non-structural elements or which are being tested. In table 1.2 the main physical and mechanical characteristics of the plastic materials are listed [42,43,44].

Table 1.1
Abbreviated terms, name and the main uses of the plastics used in the civil construction sector.

Abbreviated terms	Name	Application
PA	Polyamide	For waste landfills as a geotextile membrane [51]
PAN	Polyacrylonitrile	Fibre-reinforced concrete [52]
PC	Polycarbonate	Roofing sheets, domelights, sound walls [49]
PE	Polyethylene	Pipes, houseware, floor, soil, waste, water and gas, agricultural film [46]
PET	Polyethylene terephthalate	Fibre-reinforced concrete [53], thermal insulation [54], geotexile membrane [55]
PMMA	Polymethylmethacrylate	Profiled cladding and rooflights [50]
PP	Polypropylene	Water and gas pipes and fittings, geosynthetic turfs, wood composites, house wrap [48], fibre-reinforced concrete [53]
PS	Polystyrene	Building insulation, electrical and electronic equipment [46]
PUR	Polyurethane	Building insulation [46], insulating foams [56]
PVC	Polyvinyl chloride	Window frames, profiles, floor and wall covering, pipes, cable insulation, garden hoses, inflatable pools [46]

Table 1.2
Main physical and mechanical characteristics of the plastics used in the civil engineering.

	PA	PAN	PC	PE	PET
Density (range) (g/cm^3)	1.01-1.18 [47]	1.14-1.17 [47]	1.20-1.22 [47]	0.89-0.98 [47]	1.38-1.41 [47]
Tensile strength (range) (MPa)	82.70-90.30 [147]	372.00 [148]	62.10-65.50 [147]	4.10-60.00 [147]	48.30-72.40 [149]
Strain at break (range) (%)	15-300 [149]	17 [148]	110-150 [149]	10-1200 [149]	30-300 [149]
Young's modulus (range)(GPa)	1.58-3.80 [149]	14.60 [148]	2.38 [149]	0.17-0.28 [149]	2.80-4.10 [149]
Melting point (°C)	233 [152]	317-330 [150]	90 [153]	135 [152,153]	245 [152]

	PMMA	PP	PS	PUR	PVC
Density (range) (g/cm^3)	1.16-1.20 [47]	0.85-0.92 [47]	1.04-1.06 [47]	1.20-1.26 [47]	1.38-1.41 [47]
Tensile strength (range) (MPa)	48.00-76.00 [147]	29.00-35.00 [147]	30.00-60.00 [147]	13.79-27.58 [147]	56.50 [147]
Strain at break (range) (%)	2.0-5.5 [149]	100-600 [149]	1.2-2.5 [149]	3.0-6-0 [151]	40-80 [149]
Young's modulus (range)(GPa)	2.24-3.24 [149]	1.14-1.55 [149]	2.28-3.28 [149]	4.00 [151]	2.40-4.10 [149]
Melting point (°C)	70 [153]	170 [152]	240 [152]	220-225 [154]	163-177 [153]

1.1.3. Plastic materials used in food packaging industry

The consumption of plastics for packaging in Europe was 39.6%, as can be seen from Figure 1.8 [46]. Half of all the plastic used for packaging in Europe is destined for the food packaging, while the remaining is used in other non-food packaged products such as automotive, hardware, personal care and hygiene, cleaning and household products, medical supplies or textiles [60,61]. In Spain, in 2018, the consumption of plastic destined for packaging reached the share of 23.8% of the total plastic produced [76].

The use of plastic as a packaging material for the food sector is mainly due to the excellent qualities that it possesses: high chemical inertness, impermeability to liquids and gases, lightness, non-toxicity. Furthermore, replacing the plastic packaging with that made with another material, would lead to an increase of 300% of the weight and 150% of the volume of the packaging delivered in the waste and would increase the consumption of energy necessary for their production by 100% and therefore their cost [60]. In the food packaging sector, the polymers that are mainly used are PET, PP and PE.

If used in the form of films, which is the subject of this Doctoral Thesis, the plastic materials that are mostly used are: Low-Density Polyethylene (LDPE), Linear Low-Density Polyethylene (LLDPE), Ultra Low-Density Polyethylene (ULDPE), Medium-Density Polyethylene (MDPE), High- Density Polyethylene (HDPE), PP, PC, PVC, Polyvinylidene Chloride (PVDC), PA [62]. The plastic films that are used in the food packaging sector, since they necessarily are in contact with food, must have characteristics of safety, non-toxicity, resistance to bacteria, preserve the freshness of food, to be light and at the same time have good mechanical characteristics to protect food from damage [63]. The Figure 1.9 shows the main milestones in packaging history, from prehistoric times to the present century. Even if the first materials used for packaging date back to 10,000 BC, only since the early 1800s has the sector undergone a notable development in terms of products and materials used [89].

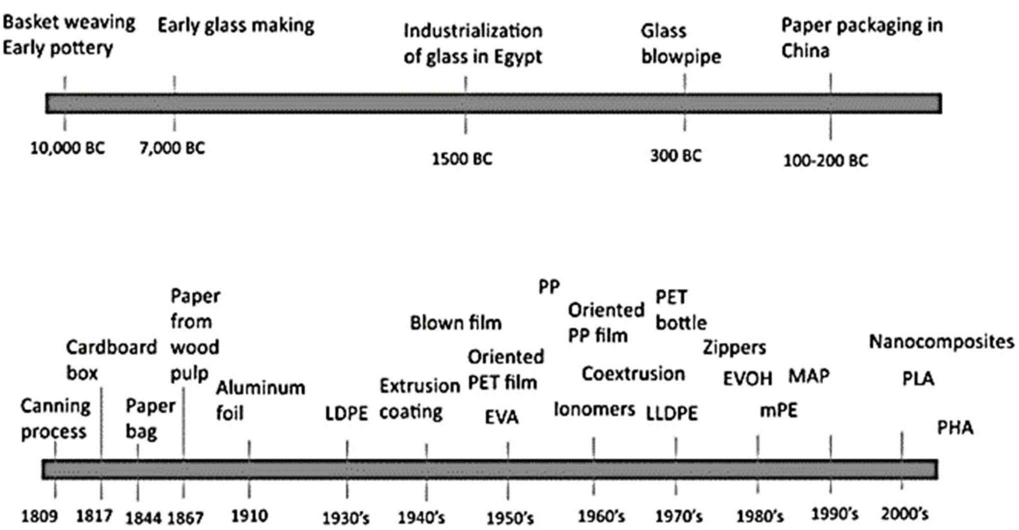
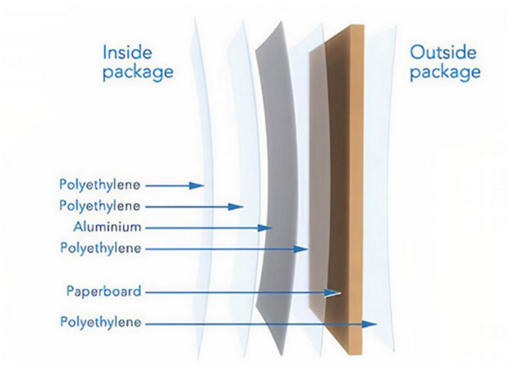


Figure 1.9 The milestones in packaging history [89].

The beginning of monolayer plastic films dates back to the early 1930s of the last century [68,69]. Only starting from the second half of the 19th century [69,70] were multilayer plastic (MLP) studied, produced and used for food packaging, made by overlapping two or more layers of different material, each intended to perform a specific task. It is a type of film that has numerous advantages over monolayers being multifunctional thanks to properties such as: reduction in weight and thickness (with the consequent economic and environmental savings in its manufacture and transport); the improvement of thermal resistance; barrier to moisture and oxygen; improvements in printing, allowing, all of them, to preserve the product they contain and extending its useful life [73].

In addition to the benefits listed above, the advantages for the industry ranging from economic, functional and aesthetic ones are not negligible [64,66,67]. Figure 1.10 shows examples of multilayer plastic for food packaging. In Figure1.10 (a) it can be seen a packaging material consisting of six layers whose materials are: the paperboard that ensures the packaging stability, strength and smoothness to the printing surface, the polyethylene that protects against outside moisture and enables the paperboard to stick to the aluminum foil and the aluminum that protects against oxygen and light to maintain the nutritional value and flavors of the food in the package in ambient temperatures [71].

Figure 1.10 (b) shows a multilayer food packaging film consisting of four layers of different materials: from inside package, Polyester (PET), Aluminum (ALU), Biaxially Oriented Polyamide (OPA STE), and Polypropylene (PP) [72]. The result obtained is that of a packaging material with high tensile strength, temperature resistance, chemical stability, reduced gas permeability and UV light protection.



(a)

(b)

Figure 1.10 Types of multilayer food packaging material structure: with six layers and three materials (Tetra Pak courtesy) (a) [71], with four layers and four materials (SP Group courtesy) (b) [72].

1.2 Problems of plastic waste

1.2.1 Legislative framework and standards related to the management of plastic waste

It is well known that plastic has now become part of all human activities and there are no sectors in which it is not presented. This has generated, around the production, use and management of the end of its life cycle, a whole series of regulations, both at European, national and regional level. The reason for this is evident as the production and use of plastic directly involves human health and, environment.

The main objectives of the most recent European legislation are to establish rules that induce society to reduce consumption and avoid the waste of plastic in addition to give rules that make it possible to clearly define the management of the end of life of plastic waste. Following the idiom "waste not, want not", and according to the report of the European Environment Agency (EEA) Report N° 02/2019 "Preventing plastic waste in Europe" [97], most European countries have adopted prevention measures on plastic residues.

The Figure 1.11 shows the "waste hierarchy" as required by the Waste Framework Directive (EU) 2018/851 [97]. Most of them are of the soft type, i.e. voluntary agreements and informative instruments. Only a few of these measures are regulatory. These measures mainly concern the production of plastic products and their consumption. The regulatory measures are mainly oriented to banning micro plastics, micro beads and some types of single use plastics and to introduce fees for plastic carrier bags [97]. Figure 1.12 summarizes the main legislation regarding packaging and packaging waste, divided into European (Figure 1.12 a) and Spanish (Figure 1.12 b).

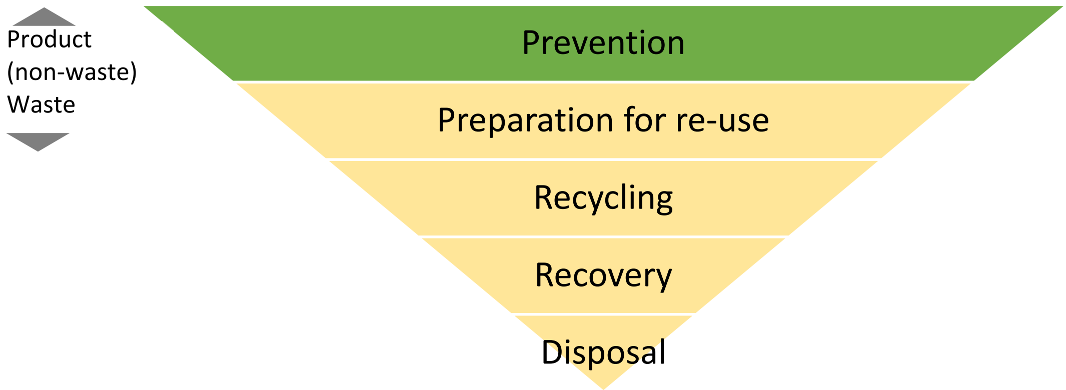


Figure 1.11 The EU waste hierarchy [97].

EUROPEAN STANDARDS

1994

Directive 94/62/EC of December 20, 1994 [98]

Subject: PACKAGING AND PACKAGING WASTE

Main objectives of the Directive:

- **Implementing some measures in order to guarantee the use of a more sustainable packaging in a future;**
- **Establishing that plastic bags cannot be free from March 2018;**
- **Prohibiting the commercialization, import and export of one-use plastic utensils from 2020;**
- **From 2030, all the commercialize packaging in EU must be reusable or easily recyclable.**

EUROPEAN STANDARDS

2004

Directive 2004/12/EC of February 11, 2004 [99]

Subject: **AMENDING DIRECTIVE 94/62/EC ON PACKAGING AND PACKAGING WASTE**

Main objectives of the Directive:

- Timescale for the transposition of directive Directive 94/62/EC into national law and set targets for the next five years;
- Clarification on the definition of "packaging" and illustrative examples;
- Encouraging the development of innovative, environmentally sound and viable recycling processes;
- Introducing cost-benefit analysis, which have indicated clear differences both in the costs and in the benefits of recycling the various packaging materials;
- Recovery and recycling of packaging waste should be further increased to reduce its environmental impact;
- The operators in the packaging chain as a whole should shoulder their shared responsibility to ensure that the environmental impact of packaging and packaging waste throughout its life cycle is reduced as far as possible.

2008

Directive 2008/98/EC of November 19, 2008 [100]

Subject: **WASTE AND REPEALING CERTAIN DIRECTIVES**

Main objectives of the Directive:

- Waste Hierarchy;
- Protection of human health and the environment;
- Principles of proximity and self-sufficiency;
- Waste Management Plans.

2015

Directive 2015/720 (EU, 2015) of April 29, 2015 [101]

Subject: **LIGHTWEIGHT PLASTIC CARRIER BAGS**

Main objectives of the Directive:

- Urging Member States to 'take measures to achieve a sustained reduction in the consumption of lightweight plastic carrier bags on their territory';
- Ensuring an annual consumption of a maximum of 90 lightweight plastic carrier bags per person by 31 December 2019 and 40 per person by 31 December 2025, and/or the levying of charges on lightweight plastic carrier bags at the point of sale of goods or products before 2019;
- Establishing reporting obligations on the annual consumption of lightweight plastic carrier bags.

EUROPEAN STANDARDS

2018

Directive (EU) 2018/851 of May 30, 2018 [102]

Subject: WASTE FRAMEWORK

Main objectives of the Directive:

- **Importance of waste prevention according to the so called waste hierarchy;**
- **Flexibility regarding the nature of waste prevention implementation;**
- **Countries waste prevention programmes and requires that objectives and qualitative or quantitative benchmarks are set;**
- **Publish periodic reports on waste prevention, every 2 years. These reports will contain a review of the progress.**

Directive (EU) 2018/852 of May 30, 2018 [103]

Subject: AMENDING DIRECTIVE 94/62/EC ON PACKAGING AND PACKAGING WASTE

Main objectives of the Directive:

- **The amendment requires Member States to take appropriate measures to encourage an increase in the share of reusable packaging placed on the market and the reuse of packaging;**
- **Minimum targets by weight for recycling will be met for specific materials in packaging waste: no later than 31st December 2025, 50% of plastic; no later than 31st December 2030, 55% of plastic.**

2019

Directive (EU) 2019/904 of June 5, 2019 [104]

Subject: REDUCTION OF THE IMPACT OF CERTAIN PLASTIC PRODUCTS ON THE ENVIRONMENT

Main objectives of the Directive:

- **Preventing and reducing the impact on the environment of certain plastic products;**
- **Promoting a transition to a circular economy by introducing a mix of measures tailored to the products covered by the directive, including an EU-wide ban on single-use plastic products whenever alternatives are available;**
- **Market restrictions (bans): the plastic products to be banned under the directive include: cutlery (forks, knives, spoons, chopsticks), plates, straws, cotton bud sticks, beverage stirrers, sticks to be attached to and to support balloons, food containers made of expanded polystyrene, products made from oxo-degradable plastic;**
- **Consumption reductions: EU countries are required to: take measures to reduce the consumption of certain single-use plastics for which there is no alternative (drinking cups including covers and lids, and containers of prepared food for immediate consumption), monitor consumption of these single-use products as well as the measures taken and report the progress made to the European Commission. The directive requires an ambitious and sustained quantitative reduction in consumption of these products by 2026 (compared to a 2022 baseline);**
- **Separating collection and designing requirements for plastic bottles: the directive sets a collection target of 90% recycling for plastic bottles by 2029 (with an interim target of 77% by 2025); these bottles should contain at least 25% recycled plastic in their manufacture by 2025 (for PET bottles), and 30% by 2030 (for all bottles);**

- **Compulsory marking: certain disposable plastic products placed on the market must carry a visible, clearly legible and indelible marking affixed to its packaging or to the product itself: sanitary items, wet wipes, tobacco products with filters, drinking cups; these labels should inform consumers about appropriate waste management options for the product or what type of waste disposal should be avoided for the product; and the presence of plastics in the product as well as the negative environmental impact of littering;**
- **Extended producer responsibility: the directive incorporates the 'polluter pays' principle.**
- **Awareness raising: EU countries must also take measures to inform consumers and to encourage responsible consumer behavior in order to reduce litter from such products, make consumers aware of reusable alternative products and the impact of inappropriate disposal of single-use plastic waste on the sewage system.**

a)

SPANISH STANDARDS

1997

Ley 11/1997
of April 24, 1997
[105]

Subject: PACKAGING AND PACKAGING WASTE

Main objectives of the Law:

- Incorporating the substantive rules of Directive 94/62/EC;
- Promoting prevention and reuse of packaging;
- Establishing the recycling and recovery targets provided for in the aforementioned Directive;
- Requirements for packaging;
- Information to be provided to the Autonomous Communities, programming and economic tools;
- The sanctioning regime.

Ley 66/1997
of December 30, 1997
[106]

Subject: AMENDMENT OF THE LEY 11/1997 OF APRIL 24

Main objectives of the Law:

- The obligations established in Chapter IV of the Law 11/1997, of April 24, on Packaging and Packaging Waste, on the "System of deposit, refund and return and integrated systems for the management of used packaging and packaging waste", will be required from May 1, 1998 and not from January 1, 1998.

SPANISH STANDARDS

1998

Real Decreto 782/1998, of April 30, 1998 [107]

Subject: APPROVAL OF THE REGULATION FOR THE DEVELOPMENT AND EXECUTION OF LEY 11/1997, OF APRIL 24, ON PACKAGING AND PACKAGING WASTE

Main objectives of the Royal Decree:

- **Definitions of packaging, composting or composting, biomethanization, economic agents responsible for packaged products, packer, first placing on the market, distance selling and automatic sale; penalty regime for packers ;**
- **Preparing business plans for the prevention of packaging waste;**
- **Recycling reduction and recovery;**
- **Obligations derived from the placing on the market of packaged products;**
- **Delivery of packaging waste and used packaging;**
- **Packaging requirements;**
- **Marking and identification of packaging;**
- **Information to Public Administrations.**

Ley 50/1998 of December 30, 1998 [108]

Subject: AMENDMENT OF THE LEY 11/1997 OF APRIL 24 ON PACKAGING AND PACKAGING WASTE

Main objectives of the Law:

- **Tax obligations for packers (se añade un apartado 4 al artículo 10 de la Ley 11/1997);**
- **Penalty regime for packers (a letter h) is added to paragraph 2 of article 19);**
- **The data on the application of the amendment will start from April 1, 1999.**

2000

Ley 14/2000 of December 29, 2000 [109]

Subject: AMENDMENT OF THE LEY 11/1997 OF APRIL 24 ON PACKAGING AND PACKAGING WASTE

Main objectives of the Law:

- **Exceptions to the application of the obligations established in Law 11/1997 in article 6 (Deposit, return and return system and integrated management systems for used packaging and packaging waste. Obligations) or, where appropriate, in the section 2nd of chapter IV (Integrated management systems for packaging waste and used packaging).**

SPANISH STANDARDS

2006

Real Decreto 252/2006, of March 3, 2006
[110]

Subject: REVIEW OF THE RECYCLING AND RECOVERY OBJECTIVES ESTABLISHED IN LEY 11/1997, OF APRIL 24, ON PACKAGING AND PACKAGING WASTE, AND BY WHICH THE REGULATION FOR ITS EXECUTION, APPROVED BY REAL DECRETO 782/1998, OF APRIL 30TH.

Main objectives of the Law:

- **Modification of the recycling and recovery targets established in Law 11/1997, of April 24, on Packaging and Packaging Waste;**
- **Modification of the Regulation for the execution of Law 11/1997, of April 24, on Packaging and Packaging Waste, approved by Royal Decree 782/1998, of April 30;**
- **Voluntary agreements and collaboration agreements between the competent public administrations and the economic agents responsible for placing packaging and packed products on the market;**
- **Establishing models related to database systems on packaging and packaging waste;**
- **Definitions in the preparation of information on packaging and packaging waste;**
- **Determination of the data;**
- **Disposal on valorization;**
- **Presentation of the data.**

2011

Ley 22/2011 of July 28, 2011
[111]

Subject: WASTE AND CONTAMINATED SOILS.

Main objectives of the Law:

- **The object, the definitions, as well as the reference to the classification and the European list of waste;**
- **Establish the legal regime for the production and management of waste, as well as the provision of measures to prevent its generation and to avoid or reduce adverse impacts on human health and the environment associated with their generation and management;**
- **Specific articles dedicated to the concepts of "by-product" and of "end of waste status" are introduced, and the conditions that a waste must meet to be considered a by-product or to lose its status as a waste are established;**
- **The principles of the waste policy: the protection of human health and the environment;**
- **A new waste hierarchy is formulated that specifies the order of priority in actions in the waste policy: prevention (in the generation of waste), preparation for reuse, recycling, other types of recovery (including energy) and, by finally, the elimination of waste;**

- **The administrative powers of each of the public administrations involved in waste management are defined;**
- **Waste management planning at national, regional and local level;**
- **The obligations of those are involved in the chain production and waste management;**
- **The transferal of waste, understood as the transport destined to elimination and valorisation**
- **The producres of elemnts that become waste are involved in the prevention and management planning of them, re-use, recycling and valorisation are encourged.;**
- **The regulation of containated soils;**
- **Responsibility, vigilance, inspection and control and sanction rules.**

(b)

Figure 1.12 The main legislation regarding packaging and packaging waste: European (a) and Spanish (b)

The reuse of waste, whatever its nature or origin, must be the goal to grant it a second life cycle. Even more so in engineering and construction sectors which accounted for 36% of final energy use and 39% of energy and process-related carbon dioxide (CO2) emissions [146]. In this framework, decarbonisation and resilience of the construction sector is necessary (responsible for almost 40% of energy and process-related emissions).

It is critical to achieve the challenge adopted on 25th September 2015 by United Nations General Assembly in Resolution 70/1: Transforming our World: the 2030 Agenda for Sustainable Development. The document lays out the 17 Sustainable Development Goals (SDG) illustrated in Figure 1.13 to end poverty and hunger, protect human rights and to protect the planet from degradation.

Figure 1.13 The 17 Sustainable Development Goals (SDGs) adopted by all United Nations Member States.

As it has been mentioned above, nowadays the massive use of plastic packaging characterized as single-use or disposable is a matter of global concern and it is one of the aspects that the 2030 Agenda develops. The relationship between the SDGs and the need to curb plastic pollution is clear. The Plastic Soup Foundation, a UN Environment Programme-accredited non-governmental organization based in the Netherlands and founded in 2011, has highlighted the relationship among several SDGs:

- SDG 3: Good health and well-being.
- SDG 6: Clean water and sanitation.
- SDG 11: Sustainable cities and communities.
- SDG 12: Responsible consumption and production.
- SDG 13: Climate action.
- SDG 14: Life below water (protection of the seas and oceans).
- SDG 15: Life on land (restore ecosystems and preserve diversity).

The reason of the massive use in packaging food sector is the excellent qualities it possesses high chemical inertness, impermeability to liquids and gases, lightness and non-toxicity. In that sense the present Doctoral Thesis is addressing an issue identified as key in the 2030 Agenda Action plan and the breakthroughs of the project advance to contribute to solve this environmental problem, not only because of the fossil resources needed to produce plastic-based films, but also because the final management mostly relies on landfill and incineration. The present Thesis will contribute to increase the recyclability rate of the plastic waste produced as it can be incorporated as reinforcement fibres for concrete mixtures in precast sector, aligning to achieve compliance with the 2030 Agenda.

At national level, the National Integrated Energy and Climate Plan 2021-2030 (PEICTI 2021-23) has identified six strategic activities:

- SA1: Health
- SA2: Culture, creativity and inclusive society
- SA3: Security for society
- SA4: Digital world, industry, space and defense
- SA5: Climate, energy and mobility
- SA6: Food, bioeconomy, natural and environmental resources

The present Doctoral Thesis is focused on achieving the SA5 strategic activity "CLIMATE, ENERGY AND MOBILITY". This challenge is defined by the National Plan for achieving a more efficient economy and society in the use of resources and water, and resistant to climate change, the protection and sustainable management of natural resources and ecosystems and a sustainable supply and use of raw materials, to meet the needs of a growing world population within the sustainable limits of the planet's natural resources and ecosystems.

1.2.2 End-of plastic waste from food packaging

The plastics market in Europe is largely oriented to packaging (39.6%), as can be seen in Figure 1.8, and, particularly, to the food market where the following polymers mainly predominate: PP (19.4%), PE - LD/PE-LLD (17.4%), PE-HD/PE-MD (12.4%), PET (7.9%) and PS + EPS (6.2%) [46].

Given the significant amount of plastic produced through the use of these polymers, which together make up 63.3 % of the plastic used in Europe in 2019 [46], the problem of the end-of-life of these products has been raised for some decades. There are three possible alternatives: recycling, energy recovery and landfilling. European statistics say that, in 2018, 42 % of the plastics used for packaging was recycled, while the rest was taken to energy recovery plants (39.5 %) and landfills (18.5 %) [46].

The objective of the European Union, according to Directive (EU) 2019/852 on Packaging and Packaging Waste, is to reach 50% in 2025 and 55% in 2030. In 2018, the Czech Republic, Germany, Spain and the Netherlands reached the goal of 50%, ahead of the time compared to the target set [46].

Recycling plastic wastes for production of secondary raw materials such as pellets, aims to give plastic a new life.

It is implemented through the following phases: collection, first sorting, shredding, washing, second sorting and control and extrusion (Figure 1.14) [46]. The first phase consists in the collection of end-of-life plastic products from separate and mix waste streams. The second is the separation of waste by type of material (plastics, aluminum, tinplate, cardboard, paper, etc.). Plastic packaging is in turn divided according to the type of polymer and by type of single or multilayer polymer [110], which will be treated. The third phase is where the plastic are ground into smaller pieces and in the fourth phase the shredded plastics are washed to remove dust and dirt. In the fifth phase, the plastics are sorted again and controlled before sent to extrusion which represents the last phase of the process during which, the plastic flakes, after being heated, are finally converted into homogenous pellets ready to use in the manufacture of new products.

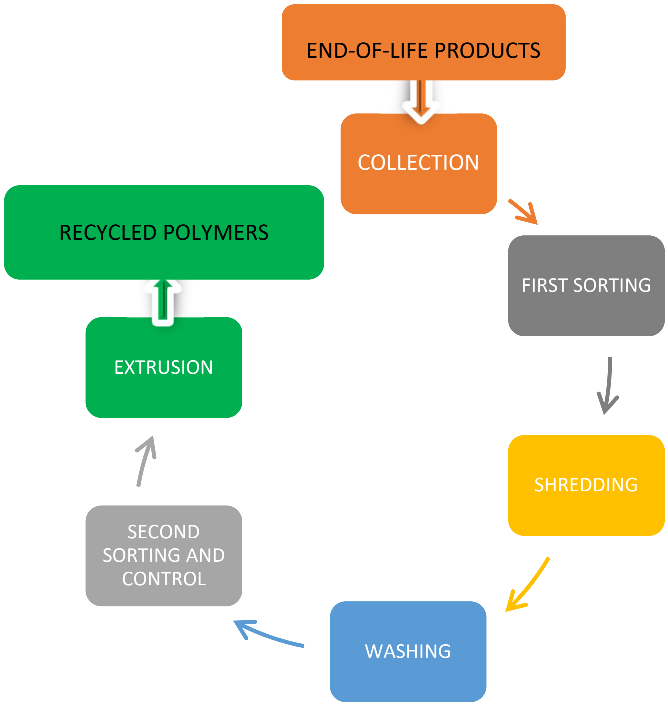


Figure 1.14 Phases of mechanical recycling of plastic (graph adapted from [46])

Another alternative to mechanical recycling is represented by chemical recycling, depolymerization, a treatment used mainly with complex plastic waste streams, like films or laminates. It consists of the chemical reversion of a polymer to its monomer or to a polymer of lower relative molecular mass and allows to obtain recycled plastics that have the same properties as the virgin ones and which therefore can also be used in applications in which there are both contact with food, as for food packaging.

The conversion process is carried out through various methods:

1. thermochemical decomposition of plastic at high temperature and in the absence of oxygen.
2. pyrolysis technology, which turns plastic waste into a secondary raw material called pyrolysis oil gasification, i.e., the creation of syngas (synthetic gas) produced by the carbon residues of the pyrolysis process, using extremely high temperatures and minimal oxygen.

3. hydrogenation, the creation of syncrude (liquid and gaseous) through the application of hydrogen in a high temperature and pressure environment.

Another option for material recovery consists of biological or organic recycling with which biodegradable plastics waste is treated microbiologically under aerobic or anaerobic conditions to produce energy in the form of heat, carbon dioxide and water vapor [115].

In addition to mechanical recycling and chemical recycling, the other option for plastics recovery technologies is energy recovery (waste-to-energy). Given the high calorific value, plastic packaging waste adapts very well to the waste-to-energy process with energy recovery and the use of this methodology would result in a significant reduction in the volume of waste going to landfills. Instead of using the primary resources of fossil fuels, energy is produced in this way in the form of generating heat, steam or electricity using plastic waste [111].

The process takes place through the incineration of the plastic and then with the total combustion of the plastic material with consequent emission of gaseous substances such as CO2, H2O, O2, N2. With modern systems and technologies, the containment of these gaseous emissions into the atmosphere is guaranteed and the combustion of plastics does not increase the emissions of harmful substances. For this purpose, the waste combustion plant is divided into five main sections: waste preparation and feeding, waste combustion, heat recovery, control of atmospheric emissions, stabilization and disposal of waste. ash and solid residues. Despite this, according to some researchers, there is evidence showing that incineration is not the terminator of plastic waste and ash and solid residues are potential sources of release of microplastics into the environment [113][114].

Incineration is the process that is mainly used for the waste-to-energy process of plastic waste that has a poor chance of being recycled otherwise, such as multilayer plastics for food packaging [116]. For these types of plastics, consisting of two or more thin layers of different materials, the recycling methods, in addition to incineration and landfill, are mainly two: the first is to separate the different components and then recycle them individually, the second is to mix all the components together in one compatibilization step (without separating into different components). Figure 1.15 shows the scheme of the recycling methods of multilayer plastic packaging materials [117].

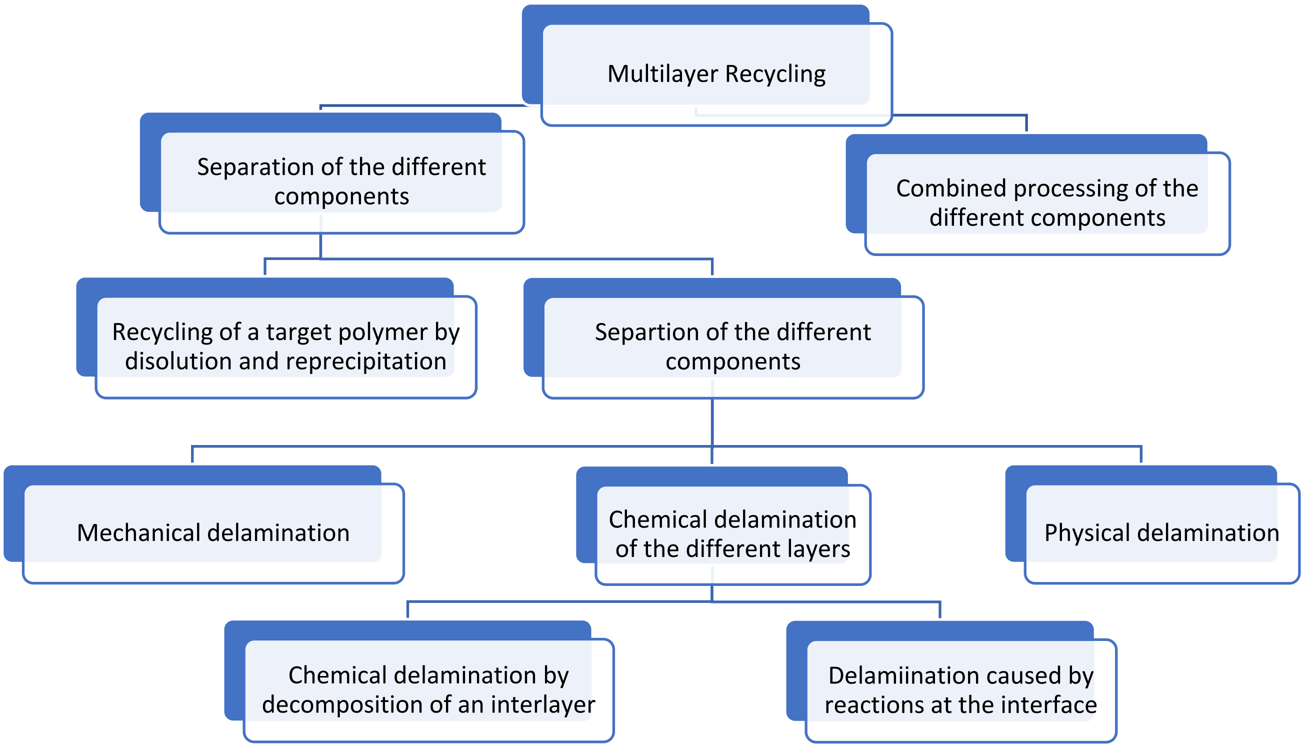


Figure 1.15 Scheme of the recycling methods of multilayer plastic packaging [117].

Helping to increase the quantity of multilayer plastic packaging waste that falls within the economic circle through its reuse is precisely the purpose of this study which, through the experimentation of innovative materials such as concrete made with the addition of macro plastic fibres obtained from industrial waste of multilayer films for food packaging, aims to present an alternative to the other recycling processes described above, which present some drawbacks regarding the environmental and energy costs.

1.3 Use of reinforcing fibres in concrete

1.3.1 Types of fibres used in reinforced concrete

As reported by multiple sources [75,90,91,92] the use of fibres as structural reinforcement in buildings dates to very ancient times, around 2500 BC, where remains of clay mixtures with straw fibres inside were found in some remains of Mesopotamian civilization dwellings.

The use of vegetable fibres as a cohesive material in the manufacture of tableware was also in use by the Incas and Maya in 2000 BC, with the aim of preventing the formation of cracks in the material placed to dry in the sun. With the introduction of the "opus caemeticium", the Romans began to add the primordial concrete with fibrous elements of vegetable or animal origin with the function of contrasting the formation of cracks and fissures. This practice was carried on almost until the beginning of the 1900s.

The search for industrial solutions that could give a satisfactory answer to the problem of cracking in the construction sector led to the use of amian and glass fibres. They were introduced in the United States around the first half of the twentieth century. Asbestos fibres were abandoned almost immediately following evidence of the damage they caused to human health. Those in glass are still used today as an additive for mortars and cement conglomerates.

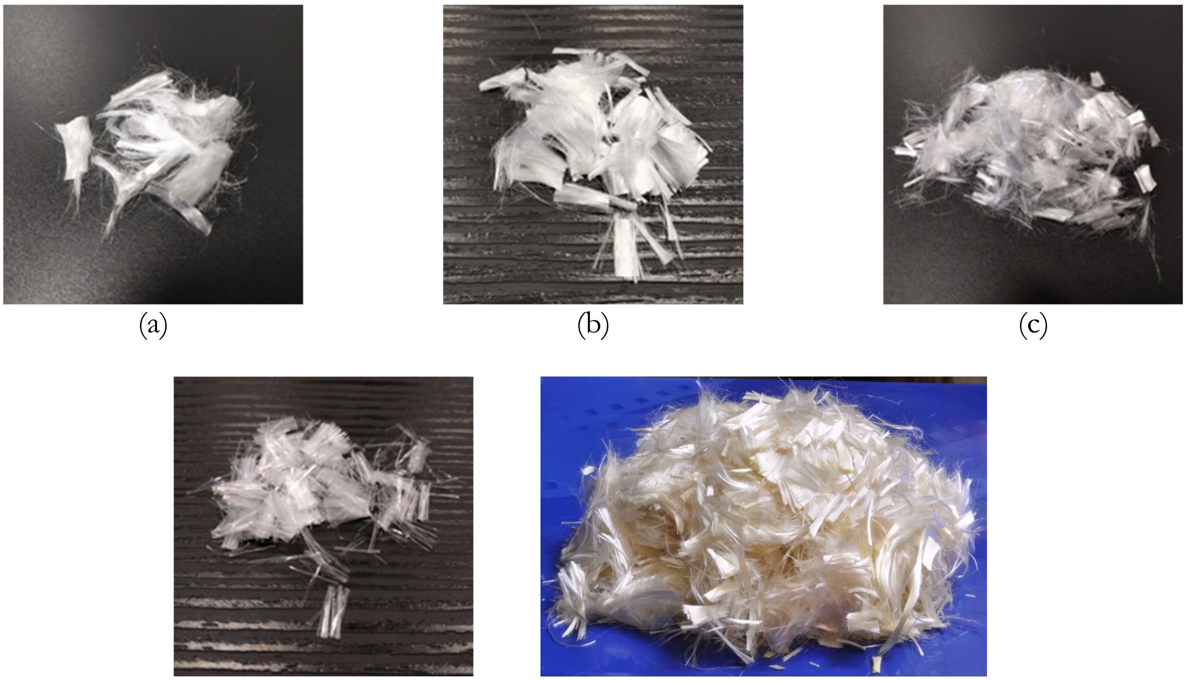
Starting in the 1960s, other types of fibres were experimented with, such as aluminum, carbon, basalt. The use of plastic fibres, as a reinforcement component of concrete for the construction of structural and not structural parts for civil engineering, began in the mid-1970s, when it became necessary to replace asbestos fibres, which proved harmful to the human health, with other types of fibres that could achieve the same benefits. The Swiss engineer Rudolf Enzler exploiting the knowledge and technologies of those times, gave rise to the use of the first polymeric fibres as a concrete reinforcement material, polypropylene (PP) [75]. Subsequently, other polymers were tested and used: the polyethylene terephthalate (PET) [77,79] the polyethylene (PE), the polyacrylonitrile (PAN) [78], the polyamide (PA) [81].

Numerous studies are still ongoing today focusing on the size, shape and quantity of polymers to be used in plastic fibre reinforced concrete. Plastic fibres are characterized not only by the type of material, but also by other geometric parameters that are: the length (distance between the ends of the fibre), the equivalent diameter (the diameter of a circle with an area equal to the average of the cross section of the fibre), the aspect ratio (the quotient between the length of the fibre and the equivalent diameter, the shape of the fibre (straight, shaped, with smooth or wavy surface). Depending on the length they are divided into micro fibres, with a variable length between 3 mm and 25 mm and mainly intended for the anti-crack reinforcement of the concrete matrix, and the macro fibres, with a length between 35 mm and 54 mm, almost always used for structural applications [82]. Table 1.3 shows the geometric characteristics and applications of the micro and macro plastic fibres most used in civil constructions [80,82,86].

Table 1.3 Geometric characteristics of the micro and macro plastic fibres most used in the civil construction sector [80,82,83,84].

Fibre types	Length (mm)	Diameter (mm)	Aspect ratio	Fibre shape	Application
Micro fibres	<25	<0.3	>200	Monofilament, fibrillated [84] multifilament [80], twisted [83]	Concrete floors, spritz-beton, prefabrication, subbase and self-leveling screeds, plasters and mortars
Macro fibres	>30	>0.3	<110	Monofilament, stranded [80] fibrillated [83], twisted [83]	Concrete floors, spritz-beton, prefabrication, slope stabilisation

In Figure 1.16 and Fig 1.17, it can be seen some types of commercial fibres used as concrete reinforcement in the civil construction sector; Figure 1.16 refers to commercial microfibres and Figure 1.17 does to macrofibres ones. It is indicated the material, the use (structural and non-structural) and the shape of the fibre.



(a) (b) (c) (d) (e)

(a) 18 mm multifilament micro fibre in polypropylene (PP) for screeds and concrete in general [87]
(b) 12 mm multifilament micro fibre in polypropylene (PP) for screeds and concrete in general [87]
(c) 6 mm multifilament micro fibre in polypropylene (PP) for lime, cement or mixed plasters [87]
(d) 12 mm fibrillated polypropylene (PP) micro fibre for screeds and concrete [87]
(e) Structural micro fibre in polyacrylonitrile (PAN) for high-performance concrete [94]

Figure 1.16 Examples of commercial micro fibres used as concrete reinforcement in the civil construction sector.

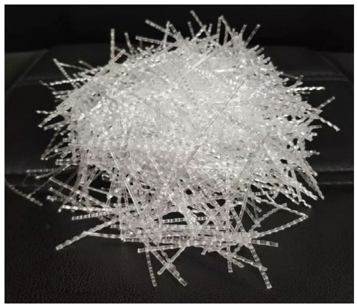

(a)

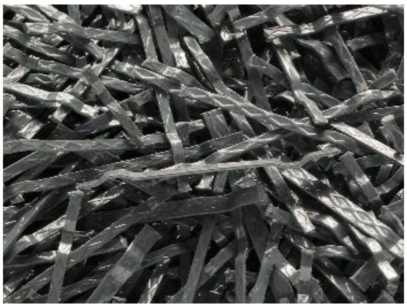

(b)


(c)


(d)


(e)


(f)


(g)

(a) Structural stranded monofilament macro fibre for industrial floors in polyolefin (PE, PP) [87]
(b) Structural stranded monofilament macro fibre for micro concrete in polyolefin (PE, PP) [87]
(c) Structural monofilament macro fibre for spritz-beton and prefabrication in polyolefin (PE, PP) [[87]
(d) Polyolefin (PE, PP) monofilament macro fibre for concrete floors [87]
(e) Structural macro fibre in polypropylene (PP) for high ductility concrete [93]
(f) Structural macro fibre in polypropylene (PP) for concrete and shotcrete [95]
(g) Macro structural fibre in polyester (PE) for projected concrete in tunnels as well as for industrial floors manufacturing and for rock walls reinforcement [96]

Figure 1.17 Examples of commercial macro fibres used as concrete reinforcement in the civil construction sector.

1.3.2. Physical and mechanical properties of concrete with plastic macro fibres

This section has been carried out by means of the research work that corresponds to the first publication named: “A state-of-the-art review on use of macro plastic fibres in concrete”, which is presented below:

A state-of-the-art review on concrete with plastic and recycled plastic macro fibres. (Publication I)

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Keywords: *sustainable construction, macro plastic fibres, circular economy, plastic waste recycling, mechanical properties, durability.*

1.3.2.1 Introduction

One of the current priority objectives of the EU is to move from a take-make-waste extractive industrial model to a circular economy. In the EU, above 25 million of tones of plastic wastes are generated per year and just 30% by weight is recycled, far from the 50% in 2025 and 55% in 2030 stablished by European [66]. This reinforces how important is to look for innovative technologies such as toconsider this stream waste as a raw-second material. Construction sector is in continuous evolution and fibres-reinforced concrete is an example of innovative solutions. The plastic fibres are useful in providing greater resistance to plastic shrinkage cracking and service-related cracking, and numerous research studies on fibresreinforced concrete have been developed [158]. The studies demonstrated that concrete made from macroplastic fibres is a composite material that could replace a part of the steel reinforcement in upstream concrete.Therefore, the exploration of the possibilities of reusing different types of plastic wastes in concrete manufacturing would lead to unbeatable results while plastics are daily consumed worldwide and the production this waste has increased immensely during the past 50 years [74].

1.3.2.2 Material and methods

The present short communication summarizes the current published literature discussing the physical andmechanical properties of concrete with plastic macro fibres. Thus, the performance of the concrete with macroplastic fibres (PF), including the recycled ones are mainly influenced by the following variables: type of plastic, dosage of plastic fibres introduced into the mixture, dimensions and shape of fibres and water to cementratio. In addition, the assessment of environmental exposure and the most important physical and mechanicalproperties of concrete containing PF: workability, air content, density, elastic modulus, compressive strength,flexural tensile strength durability have been evaluated.

1.3.2.3 Physical properties of concrete containing plastic fibres

Workability: according to various researchers, the workability, with the same water to cement ratio, depends on the amount of recycled PF used. To determine the workability of the fresh concrete was used theslump test. The results obtained were that the slump decreases with the increase in the percentage in volumeof PF [160].

Air content: most of the studies [161] indicated that the PF do not significantly affect the aircontent of reinforced concrete with PF.

*Density***:** the density of the concrete with PF has a not significative reduction respect to the conventional concrete due to the small volume of the PF added in concrete [156].

1.3.2.4 Mechanical properties of concrete containing plastic fibres

Elastic modulus: the modulus of elasticity of a concrete made with PF it is not very different from that of ordinary concrete due to the low percentage by volume of PF [160].

Compression strength: many studies have shown that the compressive strength of concrete containing PF improved upon addition of plastic fibres [85]. On the contrary, other experiments reported reductions in the compressive strength of concrete containing PF [67].

Flexural tensile strength: the studies conducted to evaluate the flexural tensile strength in concrete with PF led to the conclusion that the resistance decreases as the amount of added fibres increases [159,160]. This decrease tends to be reduced using low percentages of plastic fibres [162].

Durability: the addition of plastic fibres in the concrete, affects the durability of the mixture and, particularly, on shrinkage, porosity, and the possibility of entering gaseous or liquid substances into the concrete. Regarding the shrinkage, the results of the tests carried out by various scholars are conflicting. Someshowed a decrease in shrinkage of the concrete with PF with an increase in the fibres content [156]. On the contrary, others have seen an increase in shrinkage with the increase of the plastic aggregate [157]. The tests conducted on the porosity of the concrete contained PFs shows that the porosity increase asthe amount of PF rises [156]. According to the previous studies [85] the dosage of PFdoes not have a significant influence on the possibility of entry of gaseous or liquid substances into concrete.

1.3.2.5 Results and conclusions

The use of plastic and recycled plastic macro fibres certainly leads to positive results with regards to the protection of the environment as it significantly reduces the amount of waste taken to landfill. As it has beenshown, many of research works has been developed with the main objective of reusing recycled plastic fibresas a construction material. This research field is gaining widespread attention due to the large amount of plastic materials that society consumes daily. The conclusions of the present work are that it is feasible to revalue plastic wastes as recycled plastic fibres for being used in concrete manufacturing, improving the concrete properties. More conflicting and certainly to be continued and deepen are the research on the physicaland mechanical characteristics of the mixture between concrete and plastic fibres. In order to obtain univocalresults, thus realizing a fully usable material in the field of structural and non-structural constructions, and with characteristics equivalent or superior to concrete made only with natural aggregate. Thus, the road ahead is to continue in the investigations and experiments in this framework.

1.3.2.6 Acknowledgment

Thanks to all the authors who have carried out the studies and experiments mentioned in the present work.

1.4 Environmental impact assessment by leaching behaviour as an indicator of sustainability in concrete

1.4.1 Background of environmental assessment through leaching tests

It is widely known that the use of recycled materials and by-products in mortars or concrete as partial replacement of natural aggregate is growing interest in the engineering sector, as it reduces the demand for natural aggregate. As it has been mentioned before, scientific investigations have evaluated a wide variety of waste materials that can be incorporated to the concrete mixture: glass, coal fly ash, biomass bottom ash, aggregate from construction and demolition waste, foundry sand, tyres, iron and steel slag or plastic, the main thematic of the present Doctoral Thesis.

In recycled materials not only is necessary to evaluate the physical and mechanical properties of the waste/by-product used in the concrete, but also to assess their pollutant potential due to the presence of hazardous elements that may be released to the environment when it is put in contact at work. Then, the release level of hazardous chemical elements must be evaluated, being the leaching tests an useful analytical tool [118].

The use, recycling and final disposal of building materials constitutes an important risk to the environment. For that reason, the analysis of the potential release and the consequent migration of contaminants from the material to the environment represents one of the main objectives for assessing the environmental impact and for the sustainability of building materials. The release of contaminated substances could occur both during the initial phase of use of the material, and during use after being recycled and finally after final disposal. Contaminants, released by contact with water and transported with it, could endanger the quality of groundwater, surface and soil [119].

Leaching is the process by which soluble components (present in the tested material) are transferred from a solid matrix to an aqueous medium. Because the reproduction of this phenomenon of real scenarios is extremely expensive, laboratory tests are an indispensable tool simulating conditions that closely mimicked the actual situation. In Figure 1.18 we can see how to reproduce, by means of a laboratory test, a scenario in the field of the flow of contaminants [120].

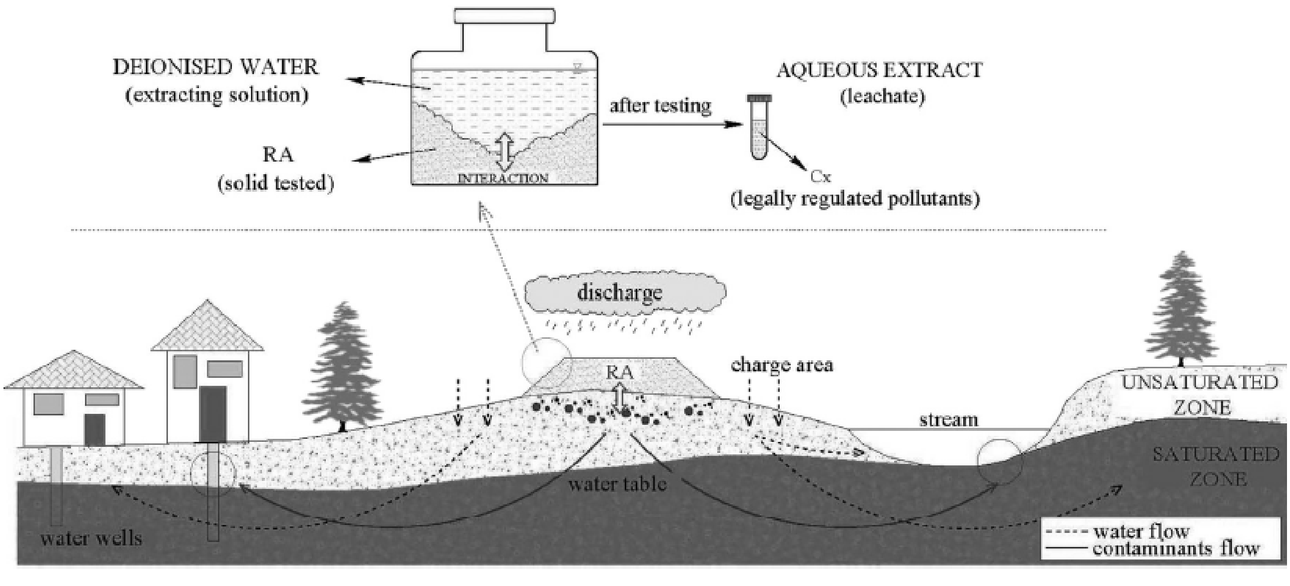

Figure 1.18 Schematic flow of contaminants in the field and extrapolation to a laboratory test [120].

Leaching tests can easily and quickly simulate the process by which constituents in a solid material (contaminated soil, sludge, sediment, compost, waste or a construction material) are released into the environment through contact with water. [139]. Understanding the rate and extent to which constituents (potential pollutant elements) of interest may be released to the leachate is central to defining the following:

1. potential environmental impacts through water-borne mechanisms including soil, groundwater and surface water contamination
2. human health and ecological risks from beneficial use and disposal of commercial materials and wastes
3. effectiveness of certain treatment processes for materials
4. designs and acceptance criteria for waste management facilities
5. degradation of structural performance of certain materials in the environment.

Another important aspect to the environmental evaluation of construction materials made from recycled products is to evaluate the parameters which affects to release levels of potential pollutant elements as:

1. chemical and physical properties of the tested material
2. chemistry of the constituents evaluated
3. characteristics of the local environment in which the material is placed, including chemical properties (e.g., pH, oxidation-reduction potential, presence of reacting constituents such as carbon dioxide) and the nature of water interaction (e.g., frequency, amount, interfacial contact area).

The present Doctoral Thesis includes the environmental behaviour of the studied material focused on leaching tests due to leaching processes allow us deep on study and research on material testing, and development of mathematical models to estimate long-term behaviour of recycled construction materials.

Numerous leaching tests are available to assess the release from these materials under a variety of conditions. In general, the most commonly used methods are percolation tests for granular materials (unbound aggregates) and diffusion tests for monolithic products (cement-treated materials as mortars or concrete) [119]. In Figure 1.19 the material flow during the environmental assessment and different leaching methodologies of a construction material, is observed:

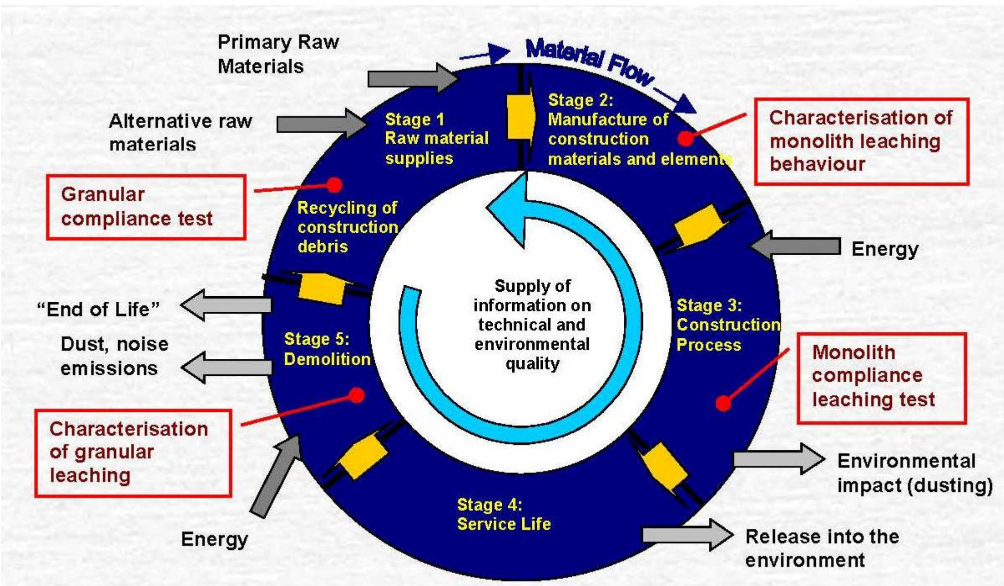

Figure 1.19 Characterisation and compliance leaching tests in different stages of the building cycle [140].

The starting point of the process observed in Figure 1.18 for construction materials made from recycled products as is the case of the material studied by the present Doctoral Thesis, is the material classification according to the pollutant potential performed according to the compliance test. The compliance test allows the classification of the waste according to its hazardous level, which conclude with the classification of this residue as inert, non-hazardous or hazardous material. If the residue complies with the legal values to be classified as inert or non-hazardous material, the following step can be achieved, and the incorporation of this waste to the dosage of the construction material (mortar or concrete) is possible. In that phase, the diffusion tank leaching tests performed to the monolithic. The different phases developed during the environmental assessment of the reinforced concrete studied are represented in Figure 1.20 following the exposed methodology:

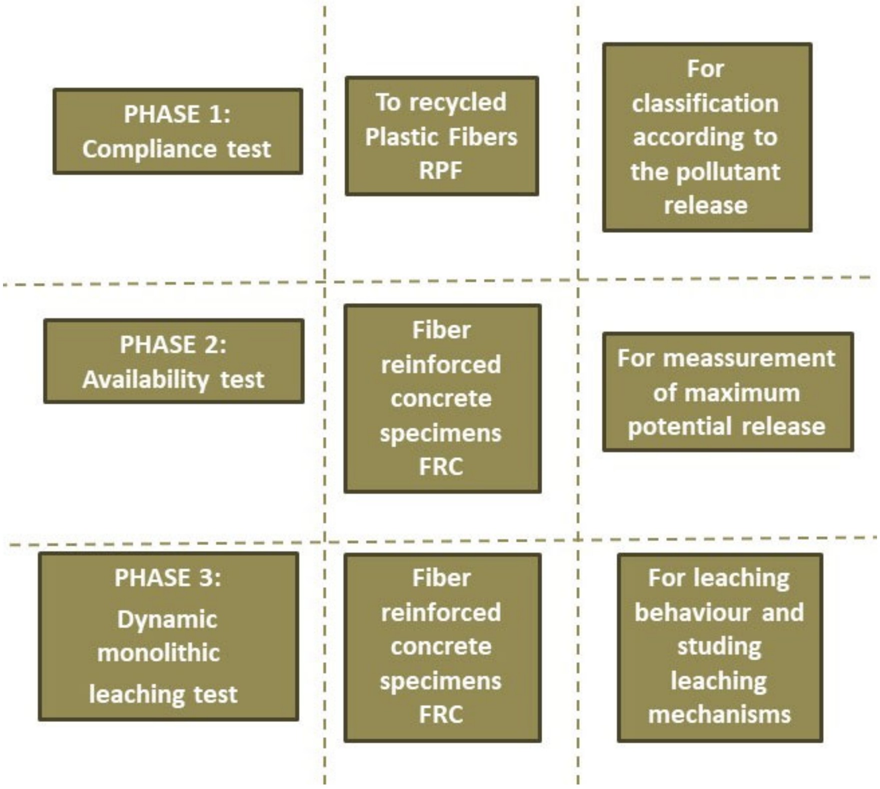

Figure 1.20 Different study phases developed during the environmental assessment of the reinforced concrete made from plastic residues.

While monolithic materials often show diffusion-controlled release, granular materials usually show percolation dominated release. Examples of monolithic products are all cementitious products (e.g., concrete, bricks, coated materials). The release of both categories (see Figure 1.21) is influenced by material specific factors as well as environmental factors.

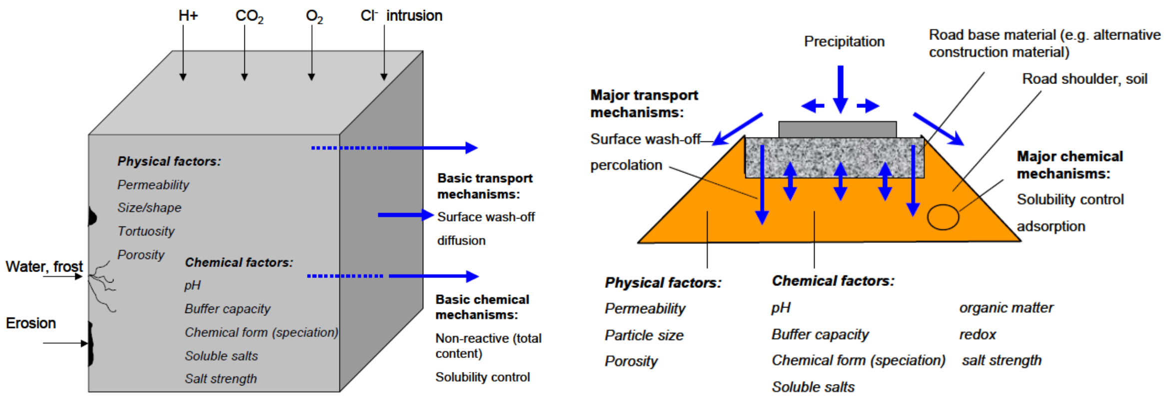

Figure 1.21 Material-specific and external factors (chemical and physical), influencing the release of contaminants from monolithic and granular material such as used in a road base [119].

Therefore, researchers focused on leaching tests as environmental tool of analysis use a common set of leaching tests that define and quantify the underlying mechanisms of contaminant release under a wide range of environmental conditions and that help us to verify that using a waste that is harmless to the environment is possible, and that it is therefore feasible, to manufacture a construction material with this by-product incorporated into the concrete dosage.

1.4.2 Leaching tests applied to characterisation of pollutant potential of reinforced concrete with macro plastic fibres

Currently the framework for leaching tests on construction products is based on a limited number of test methods suited to meet the needs of regulators and manufacturers and related to a wide range of construction materials and a wide range of application scenarios. According to that, three types of tests can be distinguished:

1. Characterization tests are procedures used as a first step of analysis. These tests characterise the release behavior of the tested material allowing a basic characterisation and reducing the number of samples that require the laboratory characterization.
2. Compliance tests allow the checking of complying with regulations in environmental matter. These tests verify whether a material complies with the behavior of a reference material. Then, after the basic characterisation, the compliance test allows to check whether the material still complies with a proper behaviour.
3. On-site verification are a type of quality control leaching tests with the aim to determine easily and quickly if a material complies with the expected behaviour for a real application (on site works for testing).

The experimental procedure in leaching tests establishes that the first step is the conformity test and to more complex and detailed characterization, the characterisation tests. In order to evaluate the environmental risk derived from the release and migration of hazardous elements that can be released using recycled materials, the release basic mechanisms must be identified.
The basic mechanisms for contaminant release of (for a wide range of environmental conditions) are:

- NEN 7371 [122,123] (maximum availability)

The Dutch standard extraction test (NEN 7371, 2004) determines the maximum leachability of an element due to the aggressive conditions imposed by this procedure (which simulates a worst-case scenario) [124, 125].

As a result, the test provides an upper limit of the leaching potential once the material is exposed in the field [126]. Thus, pH during the test is altered at very low a level which provokes a higher release of certain metals [127, 128, 121].

The Dutch leaching test NEN 7371, 2004 uses deionised water. Particles of recycled aggregate are crushed until they pass through a 125-lm sieve. The protocol consists of extracting the leachates in two steps with a liquid to solid ratio (L/S) of 50 l/kg each at pH of 7 (first extraction) and pH of 4 (second extraction). pH is kept constant by feed-back control and the addition of HNO_3. The contact time in each step is 3 h. The Dutch procedure detects the maximum leachability of contaminants from the recycled aggregate due to the restrictive conditions imposed. The test is often regarded as the worst-case scenario compared to the real situation in which a recycled material is applied in road construction.

- UNE 12457-4 [129] (compliance test)

The UNE-EN 12457 standard is a compliance test with the purpose to analyse whether the granular recycled material complies with regulations. By comparing the obtained concentrations with the legal limits indicated by the European Landfill Directive (Council Decision DC, 2003/33/EC of 19, December 2002) [130]), a material can be classified according to their pollutant potential, and the most limiting elements can be detected.

The European Standard UNE-EN 12457-3, 2002 [131] is the proposed batch test for leaching of granular waste materials and sludges at Compliance Level. The extractions are obtained in two successive stages at L/S of 2 l/kg and at L/S of 10 l/kg. The contact time with deionised water is 6 ± 0.5 and 18 ± 0.5 h respectively. In both stages, the solution is left to decant and pH, conductivity and temperature are measured. In this study, the solution was filtered through a 0.45-lm membrane filter, and subsamples of leachates from each material were collected. The samples were refrigerated until the analysis.

- UNE-EN 15863:2015 [133] (diffusion leaching tank test with periodic leachant renewal)

According to international experts, the leaching of compounds environmentally relevant from concrete samples is mainly controlled by diffusion mechanism [134, 119, 135, 136]. Consequently, the most suitable procedure for laboratory simulation of on-site behaviour of hardened concrete is a tank leaching test.

- CEN-TS 15862:2012 [137] (the single stage batch leaching test)

In the CEN-TS 15862:2012 standard, monoliths samples were contained in deionised water for 24 h with a L/A ratio of 12 cm^3/cm^2.

1.4.3 Factors that affect the contaminating potential of a material and release mechanisms

The release of potentially contaminating substances, due to the contact of construction materials with water and their subsequent migration, again through water, into the environment, could jeopardize the quality of both deep and surface waters and soil and ultimately harm human health. For this reason it is very important to evaluate what are the factors that underlie the release and transport mechanisms of potential contaminants that may be present in building materials, both in the initial phase of their use, and in the phase of their recycling and in the phase of final disposal.

- For monolithic materials (e.g., concrete, bricks, coated materials, blocks) the factors that cause the release of constituents from the materials to the aqueous phase are both of a chemical nature (pH, buffer capacity, chemical form, soluble salts, salt strength) and of a physical nature (permeability, size and shape, tortuosity, porosity).
- For granular materials (e.g., sand, sinters, gravel, steel slag) the release processes are both chemical (pH, buffer capacity, chemical form, soluble salts, organic matter, salt strength) and physical (permeability, particle size, porosity).

Two overall processes cause the release of constituents from materials to the water phase:

(1) Chemical processes (dissolution of minerals, adsorption, availability);

(2) Physical transport processes (advection, surface wash-off, and diffusion).

In practice, generally a combination of (1) and (2) cause the release to the water phase.

The basic chemical mechanisms that control the release of contaminants have been previously defined by Van der Sloot and Dijkstra, [119]:

1- dissolution of a mineral (solubility control);
2- adsorption processes (sorption control);
3- or its availability (or total content) in the product.

Some contaminants show affinity for adsorption to reactive surfaces. Positively charged heavy metal cations (e.g., Cu+2) that are not controlled by the dissolution of a mineral, are often controlled by adsorption to (negatively charged) surfaces present in the product such as organic material or oxide surfaces (sorption control). A number of inorganic constituents are not very reactive and show neither solubility control nor sorption control. Upon contact with water they will dissolve instantaneously and quantitatively. Those elements are availability controlled, as the total available concentration can be released from the product.

About the factors which affect to release, the pH of the material and that of the environment in which the material itself is located are of fundamental importance in causing the release of many constituents [119].

The pH value of the surrounding fluid determines the maximum water phase concentration at that pH value, and each material has its own pH-dependent release curve (see Figure 1.22). As it has been exposed by previous researchers, the release curves are similar and systematic for different groups of elements, only the absolute level may differ between different materials which implies that the solubility controlling phases are the same and only the relative importance of the influencing factors may differ from one material to another.

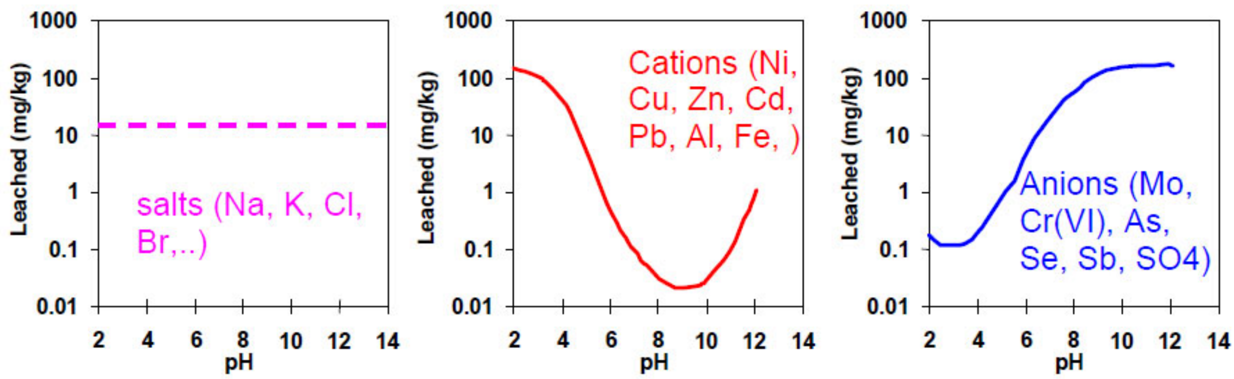

Figure 1.22 General leaching behaviour of three groups of constituents as a function of pH [119].

Aside from the chemical processes, physical transport processes determine the transfer of constituents from the material to the water phase. Three basic transport mechanisms can be distinguished:

1) advection is the process of transport of constituents taken along with the water (simulation of rain) percolating through the material. Percolation (only possible for porous materials such as granular materials) is the mechanism by which water, coming from precipitations, percolates along the tested material. Due to the further distribution of the constituent, advection plays an important role in leaching release);

2) diffusion is the transport of chemical elements solely due to the molecule's movement in absence

of flow. For that, diffusion is crucial for monolithic and compacted materials with a very low permeability and porosity. The diffusion rate is dependent on the gradient of the elements between the material and the contacting water phase.

3) surface wash-off is a process similar to advection and is a concept used to define the initial process of wash-off of soluble materials on the external surface of monolithic products. But it is important to clarify that diffusion is the major transport mechanism in monolithic materials after this initial wash-off, .

As mentioned before, it is important to distinguish granular (recycled aggregates) and monolithic products (concrete or mortars) because of the different transport regimes for these two categories. As aforementioned, the release behaviour due to contact with water is percolation dominated for granular materials and diffusion dominated for monolithic materials. As it has been stated by previous researchers [119], the amount of exposed surface area of the material increases the quantity of an element leaching from this material and diffusion proceeds faster.

1.4.4 Environmental legislation applied in this Doctoral Thesis

The considerable types of waste that are produced every year in Europe, including plastics, made it necessary to introduce specific regulations, environmental regulation, in order to preserve the quality of the environment and prevent further irreversible damage to the whole planet (to improve environmental quality). Specific environmental regulations have been issued for each of these waste categories. Those that have interested the topic of this research have mainly referred to the recycling of waste derived from plastics. The regulatory pillars referred to are basically three: the European legislation Landfill Directive 2003/33/EC [130], establishing criteria and procedures for the acceptance of waste at landfills, the Spanish legislation, the Real Decreto 646/2020, of July 7, which regulates the disposal of waste at landfills [141] and the Dutch legislation, the SQD (Soil Quality Decree) [142] containing rules with respect to the quality of soil.

The Landfill Directive 2003/33/EC [130] consists of eight articles which establish the criteria and procedures that the member states of the European Union must apply in accepting waste in landfills. The main points of the directive concern:

1. The procedure for determining the acceptability of waste in landfills.

   Each type of waste must be accompanied with a series of information necessary for it to be disposed of in landfills safely and for a long duration (type and origin, composition, consistency, leachability) also in order to understand its behaviour once delivered to the landfill (basic characterization). Subsequently, the waste must be subjected to compliance testing which will be carried out on sampling of waste according to the procedures established by the specific regulations, using the list of tests listed in the directive itself.

   Finally, the waste must be visually examined both before and after its delivery to the landfill (on-site verification), together with the related accompanying documents, in order to verify that the deposited product is the same as that which has been characterized and tested in the phases previous. Each Member State must provide for the testing requirements for on-site verification, including where appropriate rapid test methods and repeat them periodically for at least one month from the moment of acceptance of the waste in landfill.

2. The limit values and the test methods to be used for determining the acceptability of waste at landfills.

Regarding inert waste, the directive indicates a list of materials for which it is not necessary to carry out tests for their acceptance in landfills. The refusals not belonging to the indicated list must instead be subject to testing both for their acceptance and for the leaching limit values. The directive establishes the leaching limit values and the limit values for total content of organic parameters apply for waste acceptable at landfills for inert waste and granular wastes.

For monolithic waste, Member States can establish acceptance criteria such as to provide the same levels of environmental protection envisaged by the limit values relating to granular wastes. Leaching limit values are also set for non-hazardous waste, granular hazardous waste acceptable at landfills for non-hazardous waste, waste acceptable at landfills for hazardous waste. Each Member State decides which test methods to use and corresponding limit values. Finally, criteria are established for underground storage, for inert waste, non-hazardous waste and hazardous waste.

The Spanish legislation, with the Real Decreto 646/2020, of July 7 [141], represents a rule that transposes the EU directives on the landfill and environmental protection and at the same time harmonizes with the Spanish national legislation.

Its objective is to legally and technically regulate the activities of conferring waste to landfills through actions that lead to a gradual reduction of their percentage as well as to prevent, reduce and prevent harmful effects on the environment and human health from being produced. Landfills are classified according to the type of residue that is delivered to them, namely: landfills for hazardous waste, landfills for non-hazardous waste and landfills for inert waste. The waste that cannot be landfilled is also listed: those of a liquid nature, explosive, oxidizing, flammable or corrosive, infected waste, tires and all those waste that do not meet the acceptability criteria established by the decree. As regards waste that can be reused, recycled or valorised and therefore must not be taken to landfill, a specific ministerial commission will have to draw up an administrative report which will be approved by January 1, 2023 and applied starting from January 1, 2024.

The decree also establishes a whole series of rules that allow waste to be accepted in landfills only under certain conditions: having undergone preventive treatment in order to reduce the quantity deposited and the danger that these can produce for the environment and human health, control of the documentation of the residues delivered, the characterization of the waste and the related tests have been carried out for the purpose of acceptance in the landfill, the visual inspection of the residue both entering the landfill and at the point of discharge, when necessary, keep for at least three months the results of the analysis of the representative samples of the waste and the samples themselves, measure the weight of the waste.

Both in the exploitation phase and in the post-closure period of the landfill, the standard establishes that inspections must be carried out by the competent authorities, in order to ensure that the general requirements for acceptance of waste in landfill are met, all are correctly applied, the procedures and criteria for the admission of waste, the state of the landfill infrastructures and installations is assessed and that the disposal operations are free from risks for human health and the environment.

Part of the decree also deals with the management of waste associated with COVID-19 and establishes that the recommendations established by the health authorities must be followed for them.

As regards the general requirements of each type of landfill, rules are established relating to their location, water control and leachate management, soil and water protection, control of gaseous emissions, harassment and risks, the stability of the soil on which the landfill will be built and its installations and infrastructures, the safety devices that must prevent free access to the landfill site.

For the purposes of accepting waste in landfills, the decree establishes admission criteria which consist of setting limit values for leaching and limit values for the total content of organic parameters, these values contained in the tables annexed to the decree. The aforementioned limits are diversified according to whether it is a non-dangerous or dangerous residue and of a granular or monolithic type.

In addition, criteria and requirements are introduced to be able to deposit waste underground.

In the decree, a separate chapter deals with regulating the methods of sampling and testing on waste. These tests will be carried out in laboratories accredited according to the UNE-EN ISO / IEC 17025 [143] standard by the Entidad Nacional de Acreditación (ENAC), or other accreditation entities of any other member state of the European Union. The sampling, for the purposes of the basic characterization of the waste, the conformity tests and on-site verifications, will be done on the basis of the criteria established by the UNE-EN 14899: 2007 standard (Characterization of waste - Sampling of waste materials - Framework for the preparation and application of a sampling plan) [144] taking into account the technical information of the UNE-CEN / TR 15310 series [145].

The test methods on the samples, which are used to determine the general properties of the waste, the leaching tests and the analyzes, must comply with a series of rules that are listed in the decree itself.

The SQD (Soil Quality Decree), [142], is a decree, issued in 2007, by the Dutch Minister for Housing, Spatial Planning and the Environment and the State Secretary for Transport, Public Works and Water Management sustainable soil management in order to achieve a balance between the environment and man and to reconcile land use compatible with a healthy living environment. It was used in Chapter 3 of this doctoral thesis, as a reference regarding the criteria and limits of the leaching behavior of "shaped materials", including concrete with plastic macrofibres used in this research.

The decree is divided into five chapters. The first, in addition to unambiguously defining the meaning of the fundamental terms recurring in the decree itself, establishes who owns the competences, responsibilities and coordination regarding the use of construction materials, excavated soil and dredged material. The second chapter deals with the issue of quality in the execution of a job. It is pursued by introducing prohibitions, obligations and sanctions for subjects, persons or institutions, which have been previously authorized by the competent Ministries, to carry out works concerning the use of building materials and the soil.

The third chapter deals with building materials. It defines the rules concerning the composition and emission values of the substances contained in the construction material and that these values do not exceed the maximum values established and prescribed by the Ministerial authorities. The fourth chapter deals with excavated land and dredged materials used in building constructions, in road constructions, in raising land in hydraulic engineering constructions, in the redevelopment and stabilization of former quarries and mines, for the temporary storage. The uses of excavated and dredged materials containing dangerous substances are prohibited.

The standard identifies three hazard ranking systems, one concerning the specific areas of the territory where the municipal council establish local soil use values for the use of excavated soil or dredged material, one concerning standards of general use and the last concerning the works taking place on a large scale. In the last chapter, the revocations, confirmations and amendments of previous regulations are established.

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2. Objectives, Methodology and Structure of the Doctoral Thesis

2.1 Objectives

The objective of this doctoral thesis is to evaluate the potential of recycled plastic from food packaging waste as macro fibres in concrete in terms of mechanical performance and leaching behaviour. The multilayer composition of this plastic waste stream makes difficult its valorisation, as aforementioned in chapter one. The present doctoral thesis proposes the recycling of this waste as fibre reinforcement, preparing this material through cutting. This encourages the circular economy in construction and plastic sectors, to achieve the goals of the 2030 AGENDA.

Hence, this study aims to incorporate this waste as reinforcement of concrete, adding post-cracking resistance to a brittle material such as concrete. Additionally, a leaching study is required since a waste has been incorporated into a construction material. To achieve these targets, different amount of recycled plastic fibres (2 kg/m^3, 4 kg/m^3 and 6 kg/m^3) from food packaging waste were used for concrete production, as well as concrete with commercial plastic fibres and without fibres. Thus, the following points were addresses:

1. To carry out a literary review about the use of recycled plastic fibres as reinforcement in concrete with the purpose of knowing the benefits and drawbacks as well as their technical an environmental feasibility. In terms of mechanical behaviour, to compare these fibres with the commercial ones, and regarding the pollutant release, to check the different methodologies based in leaching test.
2. To evaluate the influence of the different amounts of recycled fibres used in the performance of the concretes produced, specifically in terms of:
    - physical properties (density, open porosity and capillarity);
    - post-cracking resistance; and
    - toughness and toughness index
3. To analyse the leaching performance of the new concretes produced. With this goal, some experimental methods should be carried out:
    - Compliance test for basic characterization of recycled plastic waste and its classification according to the pollutant potential.
    - Availability test for determining the maximum release level of the concrete produced.
    - Diffusion leaching tests on the new concrete produced as monoliths for, at short-term, measuring the release levels, and at long-term, pollutant behaviour of elements and identify their different release mechanisms.

2.2. Methodology

In order to achieve the aforementioned objectives, the following steps were carried out drawn in Figure 2.1. This figure highlights the relevant activities and their sequency, along with their relationship with the publications presented in this Doctoral thesis by compendium of publications.

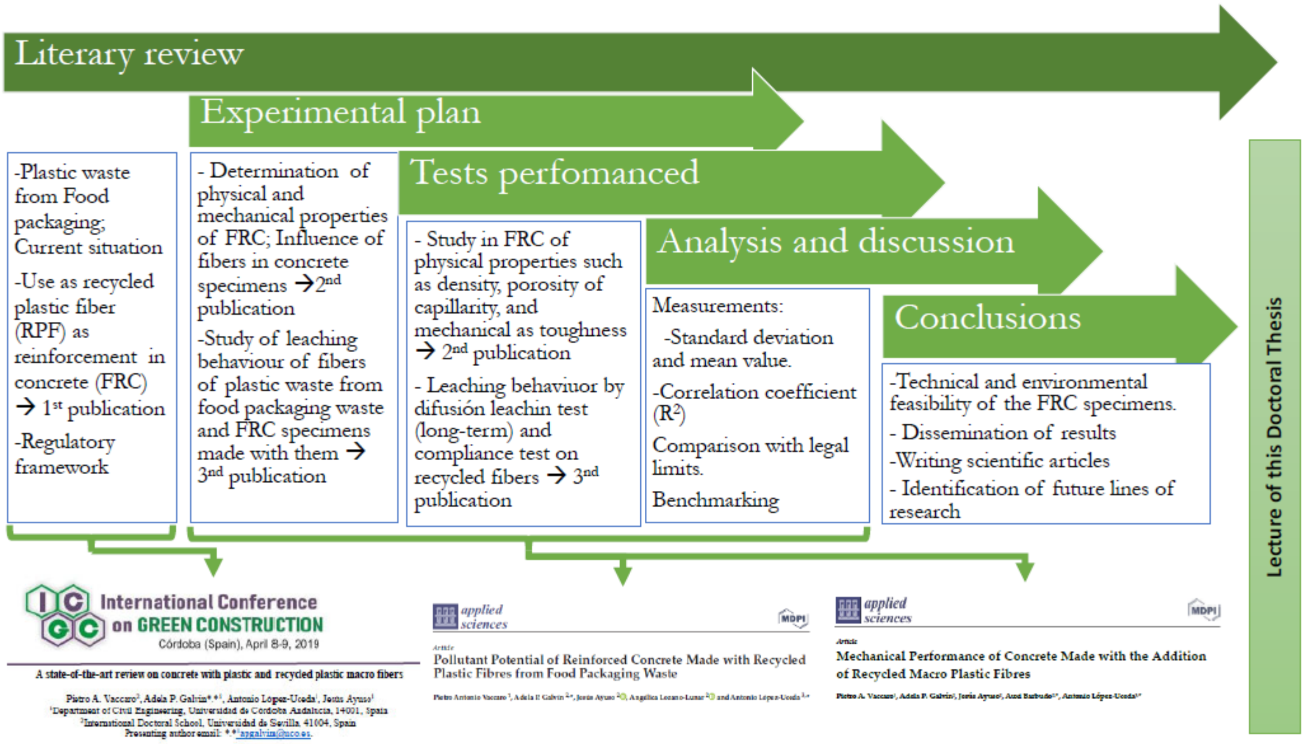

Figure 2.1 Methodology

2.3. Structure

This doctoral thesis is presented as a compendium of articles and has been structured in five chapters. Chapter 1 corresponds to the introduction. Section 1.3.2 named as "Physical and mechanical properties of concrete with plastic macro fibres" corresponds to the first publication named "A state-of-the-art review on use of macro plastic fibres in concrete" (Objective 1). The authors were: Pietro A. Vaccaro, Adela P. Galvín, Antonio López-Uceda & Jesús Ayuso. Published in NEW TRENDS IN GREEN CONSTRUCTION 53. ISBN 978-84-9927-554-3, in 2020.

The objectives, methodology and structure of the thesis are collected in chapter 2. The following two chapters (3 and 4), correspond to the two articles published in indexed international journals. These articles, in chapters 3 and 4, are published in journals belonging to the second quartile of the Journal Citation Reports.

The third chapter corresponds to the article " Mechanical Performance of Concrete Made with the Addition of Recycled Macro Plastic Fibres", the authors were: Pietro A. Vaccaro, Adela P. Galvín, Jesús Ayuso, Auxi Barbudo & Antonio López-Uceda. Published in Applied Sciences, in 2021, volume 11, number 21, page 9862. IF: 2.679 (Q2). In this study, concretes with different addition of recycled plastic fibre were produced (2 kg/m^3, 4 kg/m^3 and 6 kg/m^3); and two more with commercial plastic fibre and one without any addition. It was studied the influence of the recycled plastic fibre on the physical properties conducted such as density, open, porosity; and on the mechanical ones such as compressive and flexural strength, and toughness (Objective 2).

The fourth chapter corresponds to the article called " Pollutant potential of reinforced concrete made with recycled plastic fibres from food packaging waste" the authors were: Pietro A. Vaccaro, Adela P. Galvín, Jesús Ayuso, A. Lozano-Lunar & Antonio López-Uceda. Published in Applied Sciences, in 2021, volume 11, number 17, page 8102. IF: 2.679 (Q2). In this article, firstly, the recycled plastic fibre was tested by leaching and classified according to its potential pollutant, and secondly, at short-term and at long-term leaching tests were carried out on the concretes produced; based on the results of the second one, pollutant behaviour of elements and release mechanisms were obtained (Objective 3).

The last chapter presents the most relevant conclusions and future lines of research motivated by this doctoral thesis.

3. Mechanical Performance of Concrete Made with the Addition of Recycled Macro Plastic Fibres (Publication II)

Article

Mechanical Performance of Concrete Made with the Addition of Recycled Macro Plastic Fibres

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Abstract

For many decades, researchers have been working on finding innovative and sustainable solutions to address the enormous quantities of plastic waste that are produced every year which, after being collected, are transformed into energy, recycled, or sent to landfills. Giving a second life to plastic waste as a material to be incorporated, in the form of macro-fibres, into concrete, could be one such solution. The purpose of this study was to analyse the mechanical and physical behaviour of the hardened concrete reinforced with macro plastic fibres (RPFs) obtained from food packaging waste (FPW) discarded during the packaging phase. By varying the quantity of macro-fibres used, physical and mechanical properties such as compressive strength, modulus of elasticity, flexural strength, and toughness were evaluated. It was observed that, although the presence of macro plastic fibres reduced the mechanical resistance capacity compared to that of traditional concrete, their contribution proved to be of some importance in terms of toughness, bringing an improvement in the post-crack resistance of the composite material. This innovative mixture provides a further impulse to the circular economy.

Keywords: circular economy; recycled macro plastic fibres; physical properties; mechanical properties; toughness

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3.1. Introduction

Concrete is a widely used construction material worldwide due to its great availability of raw materials and low cost [1]. Although concrete has good compressive behaviour, it does not respond adequately under high tensile stresses. To alleviate this, steel reinforcements have usually been used to improve its tensile and flexural strengths [2–4]. However, in recent years, natural (mainly vegetable), steel, glass, and synthetic fibres have also been used [1]. Synthetic fibres help to prevent plastic shrinkage cracks in fresh concrete [5], and improve concrete performance after cracking [6].

In recent years, the EU and its member states have endowed themselves with harmonized European standards regarding the production, classification, and use of FRC (fibre-reinforced concrete) (EN 14889-1 for steel fibres, and EN 14889-2 for polymer fibres). The current trend of international legislation, therefore, is to produce a fibre-reinforced concrete (FRC) that can be classified on the basis of its guaranteed performance, and not just the volumetric percentage of fibres contained within it (EN 206:2013 + A1:2016).

Plastic fibres are synthetic fibres that are known to help improve the ductility, crack resistance, and impact resistance of conventional concrete. However, the contribution of the addition of these fibres was found to be less significant with respect to compressive strength and flexural strength [1]. Kazmi et al. [7], for their part, after investigating the fracture behaviour, mechanical characteristics, and microstructure of recycled aggregate concrete, observed an increase in mechanical properties—particularly, split tensile strength and residual flexural tensile strength—with the increase in dosage of these synthetic fibres.

Plastic fibres can be in the form of micro plastic fibres or macro plastic fibres [1]. Micro plastic fibres refer to plastic fibres with diameters ranging from 5 to 100 μm and lengths ranging from 5 to 30 mm [8]; they are used to control plastic shrinkage cracking [9], which is caused by the shrinkage of fresh concrete during the first 24 h after placement due to excessive evaporation of bleed water [10]. Macro plastic fibres, which were used in this study, normally have a length of 30–60 mm and a cross-section of 0.6–1 mm^2 [11]; these are used not only to control plastic shrinkage [12], but also to control drying shrinkage [6]. Another significant benefit is the post-cracking performance provided by the addition of macro plastic fibres [13,14].

On the other hand, world plastic production reached almost 368 million tons in 2019, of which approximately 58 million tons was generated in the EU (European Union). Of all the plastic produced in the EU in the year 2019, more than 20% was used in the building and construction sector, while almost 40% was used in packaging [15].

In the EU, in 2018, 24.9% of collected plastic waste was landfilled, 42.6% was transformed into energy (by incineration [16]), and the remaining 32.5% was intended for recycling [15]. According to a recent report [16], the target set by the new EU Directive 2019/852 provides for the achievement of a recycling percentage of plastic for packaging of 50% by 2025, and 55% by 2030. Hence, plastic wastes are universally considered to be a threat to the environment [17].

Among the various types of recycling management approaches, the reuse of waste and recycled plastic material in the construction industry is considered an ideal method for disposing of plastic waste [18]. A review has been published recently about current trends in plastic waste composites as construction materials [19], reporting diverse uses of the most common plastics, such as polypropylene (PP), polyethylene (PE), and polyethylene terephthalate (PET), after proper recycling treatment—for instance, in bricks, as aggregate or filler in concrete, or as reinforcement in concrete in paving tiles or slabs. Benefits and drawbacks are weighed, highlighting as benefits the fact that its use increases its value, so recycling is encouraged, and raw material consumption is reduced; meanwhile, as drawbacks, sorting and selection, and the lack of study of recycled plastic waste in construction materials, are cited. Other studies, such as the one carried out by Platon et al. [20], dealt with other uses, such as the production of composite materials for sound insulation purposes produced by thermocompacting mixed plastic waste, from the production process of sanitary wares, and thermoplastic polymers.

The use of recycled aggregates from plastic waste to replace natural aggregates has been previously studied. Mohammadinia et al. [21] showed the feasibility of substituting up to 50% of natural aggregates for a combination of recycled plastic and glass aggregates in the construction of concrete footpaths. Corbu et al. [22] found that a combination of recycled aggregates from waste plastic and waste glass can totally replace natural aggregates in precast concrete pieces. Thorneycroft et al. [23] proved the feasibility of substituting up to 10% by volume of sand for recycled plastics in the manufacture of concrete. Elsewhere, Merlo et al. [24] detected a lack of adhesion between recycled plastic and cementitious matrix, explaining the loss of mechanical properties as ratio incorporation increased.

In addition, authors such as Foti went even further, studying the use of PET plastic waste in strips of large size [25] or in grids of macro-strips [26] as reinforcements in concrete, suggesting the possibility of being considered for the reinforcement of concrete in place of steel. Within this sector, recycled plastic fibres can be used in FRC, which allows improvement of the concrete properties, in addition to the environmental benefits that this entails [4,18,27,28].

The global value of the packaging industry is around EUR 345 million, of which the EU accounts for one-third, and of which 50% is related to food packaging. Forecasts indicate that the sector will continue to grow both in size and in importance [29]. In terms of structure, monolayer and multilayer can be distinguished: monolayer films consist of a thermoplastic polymer sheet made mostly of PE, PP, or PET, with a range from 20 to 200 μm of thickness, while multilayer films are made up of a different number of sheets that can be polymeric (thermoplastics) or non-polymeric materials (paper or aluminium foils) [30]. Multilayer plastic waste used in food packaging does not enter the waste recycling stream due to the wide variety of materials, its multilayer composition [31], and the colours that make up the food packaging; hence, a call for a circular economy is proposed [32]. Currently, the technical feasibility of recycling this waste at low cost is not close to being achieved [31,33]. Indeed, due to the fact that it is not always managed properly, the recycling process is quite complex and, with the great environmental concern attached, the plastic sector is encouraged to participate in projects to deal with this issue [34,35].

This confirms the need to address studies such as the present work, which are essential in finding solutions that transform plastic waste from packaging into new resources, enabling not only contribution to the circular economy, but also the production of innovative materials for civil construction.

In particular, this study deals with the analysis of the mechanical behaviour of concrete with the addition of macro-fibres obtained by cutting plastic sheets; specifically, measurements such as crack mouth opening displacement (CMOD) and toughness were obtained. Three different levels of fibre addition were used in the production of concrete (2 kg/m^3, 4 kg/m^3, and 6 kg/m^3), and two reference concretes were produced (one without fibre, and another with a commercial fibre). The main novelty of this research consists of having used not commercial plastic fibres, but waste material derived from the processing of plastic for food packaging. These multilayer plastic wastes are difficult to recycle due to their heterogeneous composition. Hence, this work aims to valorise this waste, contributing to a circular economy.

3.2. Materials and Methods

3.2.1. Recycled Plastic Fibres (RPFs) from Food Packaging Waste (FPW)

The recycled plastic waste (RPW) consisted of macro plastic fibres obtained from the waste derived from the production and processing of plastic, supplied in waste rolls by a local company that deals with the production of multilayer plastic for packaging. The plastic sheet was made of four overlapping layers of different materials: polyester, aluminium, biaxially oriented polyamide, and polypropylene (PET + ALU + OPA STE + PP G). The fibres used in the experimentation were obtained in the laboratory by cutting plastic sheets (Figure 3.1a,b). A commercial polypropylene fibre (CPF) was also used as a comparison (Figure 3.1.c). The fibres were in compliance with the EN 14889-2:2006 standard. The dimensions and main properties of the plastic fibres used are summarised in Table 3.1.

(**a**) (**b**) (**c**)

Figure 3.1. Plastic fibres: plastic waste rolls from food packaging (**a**); the prepared fibres (**b**); commercial polypropylene fibre (**c**).

Table 3.1. Technical data of recycled plastic fibres (RPFs) and commercial polypropylene fibre (CPF).

Technical Data	Unit	RPF	CPF
Thickness	mm	0.125	-
Width	mm	4	-
Length	mm	50	50
Diameter	mm	-	0.682
Aspect ratio	-	63.00	73.53
Density	g/cm^3	1.13	0.91
Tensile strength	MPa	500	530
Modulus of elasticity	GPa	9.0	7.4

3.2.2. Fibre-Reinforced Concrete (FRC) Specimens Made with RPF

Specimens of PFRC were prepared by adding recycled plastic fibres (RPFs) to the concrete mix at 2, 4, and 6 kg per m^3 of concrete. The prepared mixtures were assigned the following codes: PFRC-2, PFRC-4, and PFRC-6, respectively. Two control specimens were also prepared: a control specimen of concrete without reinforcement (C-REF), and fibre-reinforced concrete made with a commercial polypropylene fibre (PFRC-REF; dosage of 2 kg per m^3 of concrete).

The aggregates used for manufacturing the concrete mixes in the laboratory were coarse gravel (7–25 mm) at 960 kg per m^3 and sand (0–7 mm) at 970 kg per m^3, both with a siliceous nature. A total of 280 kg of CEM II 42.5 AV-R cement per m^3 of concrete produced was used, reaching a water-to-cement ratio of 0.54. In order to improve the workability and avoid segregation of the mixture, along with having an S3 slump class according to UNE-EN 206:2013 + A1:2018, two different types of additives were used: the plasticizer Complast MR260 at 5 mL per kg of cement, and the superplasticizer Structuro 357 at 7.7 mL per kg of cement. The slump ranged between 15 cm for C-REF and 10 cm for PFRC-6 mix. Slump values decreased as macro-fibre content increased, in agreement with the findings of other authors [18]. The concrete produced for each single mixture was casted, resulting in nine cubic specimens with a length of 100 mm, three prismatic beams measuring 100 mm × 100 mm × 400 mm, and six cylindrical specimens with a diameter of 150 mm and a height of 300 mm, which were then cured in a climatic chamber (at 20 °C and 95% ± 5% relative humidity) and subjected to testing at various ages.

3.3 Experimental Methods

3.3.1. Physical Properties

The dry density and the open porosity of the concrete specimens were obtained following the standard UNE 83980:2014. The performance of the mix concrete in terms of capillarity was determined using the Fagerlund method, following the standard UNE 83982:2008. Both tests were carried out on three cubic specimens with a length of 100 mm.

3.3.2. Compressive Strength

The compressive strength test was performed in accordance with the UNE-EN 12350-2:2009 standard; it was carried out on three cubic specimens with a length of 100 mm after 7 and 28 days of curing, as well as on three cylindrical specimens with a height of 300 mm and a diameter of 150 mm after 28 days of curing.

3.3.3. Elastic Modulus

The elastic modulus was determined, in accordance with UNE-EN 12390-13:2014, after 28 days of curing on three cylindrical specimens (Ø150 × 300 mm).

3.3.4. Flexural Strength

The flexural strength was determined using three prismatic specimens (400 × 100 × 100 mm^3 by length × width × height) after 28 days of curing. The tests were carried out using a four-point bending tensile testing machine, with notch-opening control, complying with the UNE 83509:2004 standard.

3.3.5. CMOD and Toughness

The toughness index and CMOD were determined for each mixture by subjecting three prismatic specimens (equal to the flexural strength ones), notched (5 mm in width and 20 mm in height) in the central part, to a four-point bending tensile test, after 28 days of curing, in accordance with the UNE 83510:2004 standard.
The measurement of the crack opening at the mouth of the notch of the specimen (CMOD) was performed using two linear variable displacement transducers (LVDTs) attached to the bottom of the concrete specimen. At the end of the test, the fissure was unique and corresponded to the notch. During the test, the applied loads and the corresponding opening of the notch made along the centre of the specimen were detected and recorded, thus obtaining, for each mix design, the load–CMOD curve. From the load–CMOD curve, the load and the relative resistance at the point of first cracking of the specimen were determined. The values of toughness T (N·m) were obtained by calculating the area under the load–deflection curve, from the point where the linear section of the diagram began, and up to the point where the arrow reached the value of 1/150 of the span between the supports of the specimen. The toughness index, I_{30}, was obtained as the ratio between the area under the curve from the origin of the curve to the point where the deflection reached the value of 15.5 times the deflection corresponding to the formation of the first crack, and the area under the curve between the origin of the curve and the deflection related to the first crack.

3.4. Results and Discussion

3.4.1. Physical Properties

The results of the dry density and water absorption in concrete samples are depicted in Table 3.2. The dry density values ranged between 2.21 g/cm^3 and 2.25 g/cm^3 for mixtures with recycled fibres, and between 2.26 and 2.27 g/cm^3 for reference concretes. Thus, owing to the small volume fraction of the plastic fibre added to the concrete, there was no significant reduction in the density of plastic FRC as compared to the density of conventional concrete, in accordance with the findings of de Oliveira and Castro-Gomes [36], Gu and Ozbakkaloglu [18], Han et al. [37], Karahan and Otis [38], and Richardson [39].

The open porosity (Table 3.2) of concretes with recycled fibres presented values between 12.17% and 13.34%, compared to between 11.84% and 14.15% for reference concretes. In all cases, the concretes made with fibres presented higher porosity values than the concrete made without them (C-REF). However, mixtures with recycled fibres presented lower values than those obtained for concrete made with commercial polypropylene fibres (PRFC-REF).

Even the effective porosity of concrete, with values ranging from 5.48 × 10^{-2} cm^3/cm^3 to 8.72 × 10^{-2} cm^3/cm^3 (corresponding to the reference mixture), did not undergo significant variation.

The resistance to water penetration by capillarity absorption, and the capillary absorption coefficient—with values ranging between 24.98 min/cm^2 and 34.64 min/cm^2 and between 9.51 × 10^{-2} kg/(m^2·min$^{1/2}$) and 14.81 × 10^{-2} kg/(m^2·min$^{1/2}$), respectively—highlighted a slight decline in resistance to water penetration by capillarity absorption, and a slight increase in the capillary absorption coefficient compared to the reference values. This is consistent with the studies carried out by other researchers [40,41], who reported that the addition of plastic fibres to concrete does not affect the properties studied, since the values obtained differ little from those of the fibre-free concrete mix.

Karahan and Atis [38], on the other hand, indicated that the water porosity and the water absorption capacity of concrete containing PP fibres (with concentrations lower than 0.20%) increased compared to the corresponding value for the control mix.

Table 3.2. Results of the test methods conducted on concrete samples, related to their physical properties.

Test Methods\Mixes	C-REF	PFRC-REF	PFRC-2	PFRC-4	PFRC-6
Dry density (g/cm^3)	2.27 (0.01)	2.26 (0.01)	2.24 (0.01)	2.25 (0.01)	2.21 (0.02)
Open porosity (%)	11.84 (0.24)	14.15 (0.31)	12.21 (0.20)	12.17 (0.44)	13.34 (0.49)
Effective porosity of concrete, ε_e (×10^{-2} cm^3/cm^3)	5.48 (0.38)	8.72 (0.65)	6.30 (0.20)	6.55 (0.08)	6.53 (0.07)
Resistance to water penetration by capillarity absorption, m (min/cm^2)	33.22 (1.01)	34.64 (4.70)	27.62 (0.62)	28.39 (1.79)	24.98 (0.90)
Capillary absorption coefficient, K (×10^{-2} kg/(m^2·min$^{1/2}$))	9.51 (0.50)	14.81 (0.60)	11.98 (0.44)	12.30 (0.27)	13.08 (0.37)
The standard deviation is shown in parentheses					

3.4.2. Compressive Strength

As can be seen from the graphs in Figure 3.2, the addition of RPF to the mixture reduced the compressive strength at 7 days and 28 days in the cubic specimens, as well as in the cylindrical specimens at 28 days. This reduction increased with the increase in the amount of fibre added. The percentage variations in the compressive strength, as a function of the type and quantity of fibre, compared to the control specimen (C-REF), exhibited, for the cubic specimens at 7 days of curing, decreases of 16.66% for PFRC-REF, 31.72% for PFRC-2, 32.68% for PFRC-4, and 31.62% for PFRC-6. For the cubic specimens, at 28 days of curing, the corresponding decreases were 15.95% for PFRC-REF, 20.73% for PFRC-2, 23.98% for PFRC-4, and 26.28% for PFCR-6. From these data, it can be seen that the compressive strength, in the cubic specimens, underwent a minor reduction compared to C-REF, with an increase in curing time. For the cylindrical specimens, at 28 days of curing, the corresponding decreases were 18.39% for PFRC-2, 36.78% for PFRC-2, 47.89% for PFRC-4, and 54.38% for PFRC-6.

For the cylindrical specimens, there was a greater reduction in compressive strength than recorded for the cubic specimens. This behaviour could be attributed to the different manufacturing methods of the two types of specimens, i.e., the vibrating method for cubic specimens and tamping with a steel rod for cylindrical specimens. Furthermore, the similar compressive strength performance presented by cubic specimens with RPF and CPF can be highlighted.

In general, a reduction in compressive strength with an increase in RPF content occurred, consistent with the investigations of Kim et al. [42] and Meddah and Bencheikh [29]. This could, according to some authors, have been due to the poor homogeneity and compactness of the mixture [43,44], or to other factors such as (1) the formation of zones inside the mixture where stress concentrations are created that favour the propagation of damage, (2) a weak connection at the concrete–plastic fibre interface, (3) an increase in air content [45], and (4) the presence of further voids due to the fibres [4,46].

However, other researchers—such as de Oliveira and Castro-Gomes [36], Han et al. [37], Hsie et al. [47], Gu and Ozbakkaloglu [18], and Kakooei et al. [48]—reported the opposite, i.e., that the compressive strength of the concrete improved upon the addition of plastic fibre.

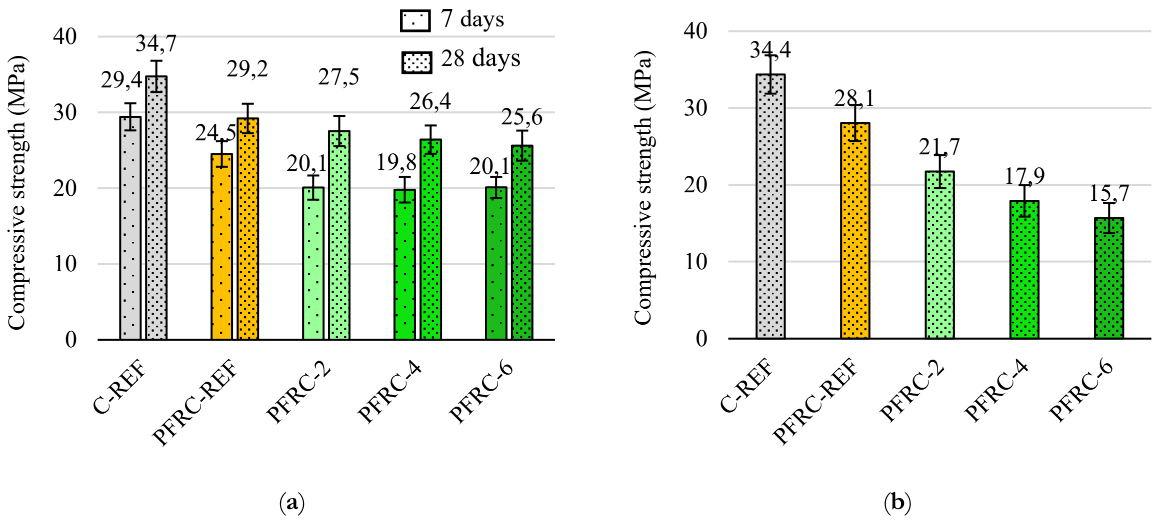

Figure 3.2. Compressive strength values for each mixture, along with the standard deviation bands: at 7 and 28 days ($f_{ck,cube}$) (**a**); at 28 days ($f_{ck,cyl}$) (**b**).

3.4.3. Elastic Modulus

Figure 3.3 shows the variation in the values of elastic modulus (E_{cm}) with the quantity of recycled plastic fibres. There is no consensus in the literature with respect to this property. Kim et al. [42] reported that, for all mixtures containing recycled plastic fibres, a decrease in E_{cm} could be noted with respect to the values obtained in the control specimen. Other researchers observed an increase in E_{cm} with an increase in the percentage of plastic fibre [4,49], whereas others—such as Pelisser et al. [50], Karahan and Atis

[38], and Mazaheripour et al. [51]—indicated that the E_{cm} of the plastic FRC did not differ significantly from that of conventional concrete. Our results (Figure 3.3) were consistent with this latter finding, since the mixtures made with plastic fibres showed very similar values, except for PFRC-2, which showed an increase of 31% with respect to its reference counterpart (PFRC-REF). This could be attributed to the greater modulus of elasticity presented by the FRP than the CPF, as seen in Table 3.1.

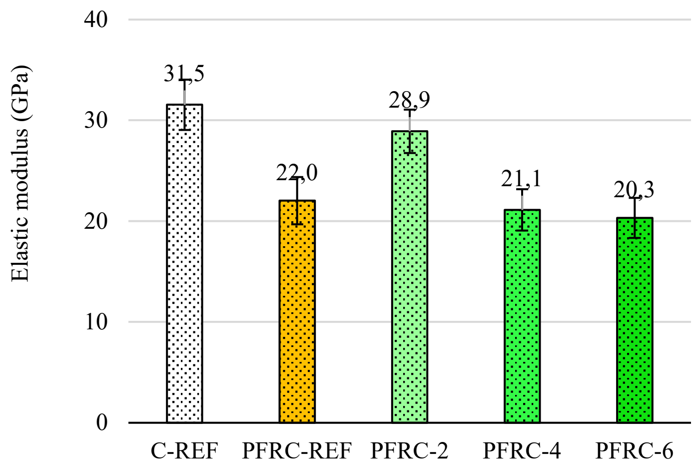

Figure 3.3. Elastic modulus (E_{cm}) values for all mixtures at 28 days, along with the standard deviation bands.

3.4.3. Flexural strength

Most previous studies observed that the flexural strength of concrete increased upon the addition of PF [36,50–54].

However, as can be seen in Figure 3.4, the flexural strength recorded for non-notched prismatic specimens underwent a considerable reduction for mixtures made with fibres compared to the control concrete specimen (C-REF). The results also show that, as the percentage of fibre increased, a significant reduction in flexural strength followed. This is consistent with the findings of other authors [55,56], who stated that the flexural strength only improved when the concrete had a low fibre content, whereas, above a certain threshold, the flexural strength decreased (which coincides with our case).

With respect to the values of the control concrete specimen (C-REF), the decreases were 28.45% for PFRC-REF, 39.79% for PFRC-2, 45.03% for PFRC-4, and 49.04% for PFRC-6. According to other researchers, the reduction in flexural strength may have been due to the presence of voids inside the concrete matrix [46], which depends mainly on the strength of the mix, as seen in concrete specimens with fibres compared to C-REF.

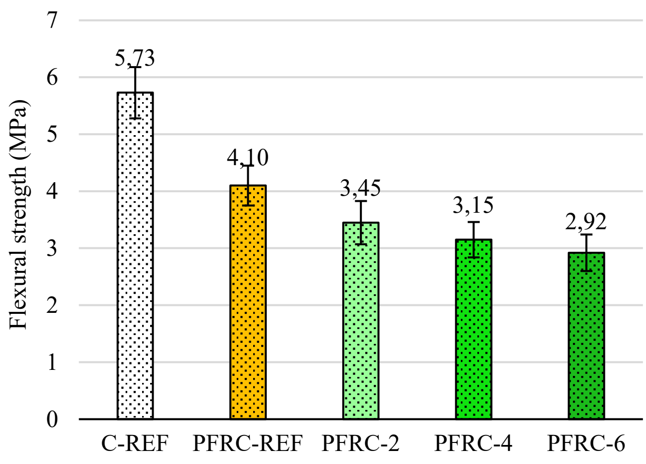

Figure 3.4. Flexural strength values for all mixtures, along with the standard deviation bands.

3.4.5. Toughness

The experimental curves of the relationship between the applied load (F) and the CMOD as a function of the type and quantity of fibre used in the mixture are presented in Figure 3.5, where the punctual values of the crack opening are shown on the *x*-axis. After reaching the point of first cracking, there was a sudden drop in load, probably caused by both the failure of the anchoring of some fibres and the breakage of others. In correspondence with the broken surface of the specimen, slipped and broken fibres were present. The post-cracking phase followed, during which the plastic fibres began to make their contribution and the deformation energy was dissipated by the fibres with the consequent remaining load, which remained constant up to a CMOD of nearly 4.5 mm, as depicted in Figure 3.5. This same behaviour was observed by many other researchers using different types of commercial or recycled plastic, and with different amounts of plastic fibre [14,41,44,57].

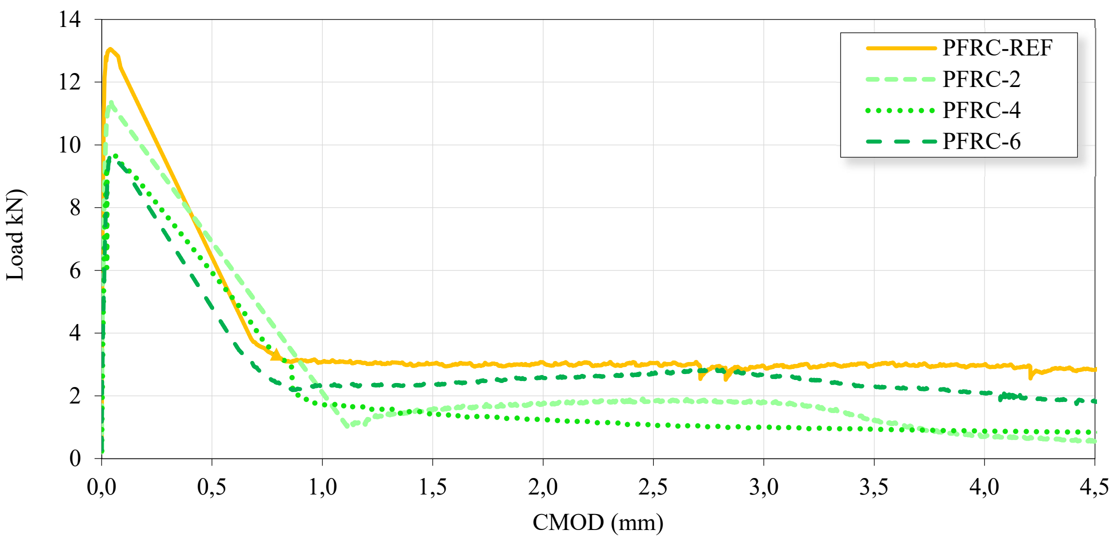

Figure 3.5. Average load–CMOD curves of three specimens of each concrete type.

Figure 3.6a shows the load–deflection graphs, while Figure 3.6b shows the initial segments of the graphs obtained by linearly interpolating the forces between the values of 20% and 60% of the peak force, where the first break occurred, as dictated by the UNE 83510: 2004 standard, which allowed us to calculate the toughness and the toughness index. The behaviour of the four mixtures was generally similar, wherein there was a first phase where the force increased until it reached its maximum value (peak value), and where the first crack occurred in the specimen. With regard to the mixture made with the addition of commercial plastic fibres, the graph displayed a value of peak force greater than that of the other three mixtures. Subsequently, there was a rapid decrease in strength, followed by a slight increase, which may have been due to the contribution of the plastic fibres contained within the mixture. Toward the end, the force gradually decreased until the specimen broke, showing a degrading type of post-cracking behaviour (softening behaviour). The same results were obtained by numerous other researchers who experimented with the addition of recycled plastic fibre to reinforce concrete, demonstrating that it is possible to obtain a concrete that has good mechanical characteristics in terms of toughness, as well as reduces environmental pollution by reusing material resources that would otherwise be disposed of in landfills [12,27,58,59]. In the post-cracking phase, the concrete mix with the highest fibre content (PFRC-6) exhibited a similar behaviour to the concrete mix with commercial fibre (PFRC-REF).

Figure 3.7 shows that the presence of plastic fibres inside the matrix increased both the ability of the concrete to resist the advancement of cracks, and its ability to absorb energy and deform plastically before breaking, giving the compound a certain residual tensile strength after the microcracking of the cement matrix. In Figure 3.7a, it can be seen how the toughness of the concrete with commercial fibre (PFRC-REF) was higher than that of the concrete with recycled fibre (PFRC-2) when using the same quantity of fibre. This was mainly due to the commercial fibre used in this study having a much rougher surface than the surface of the recycled fibre, which was practically smooth. The increase in toughness index (Figure 3.7b) resulted in the value of the strength index of the mixture with no fibre addition (C-REF) being equal to 1. An increase in the percentage of recycled fibre confirmed the stitching action of the fibres themselves inside the cement matrix, which was fundamentally dependent on the type of fibre and its quantity. Comparing the RPF concrete specimens with the reference (PFRC-REF), the toughness index increased with RPF dosage, reaching similar levels at 6 kg of RPF per m^3 of concrete.

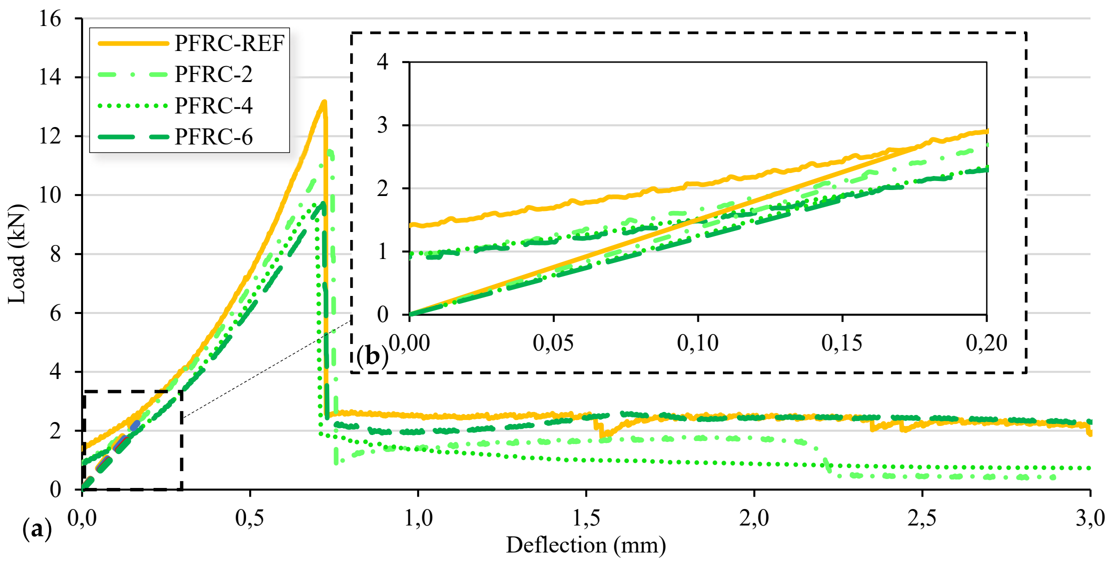

Figure 3.6. Load–deflection curves of each concrete type (**a**); detailed view of the initial section of the curve (**b**).

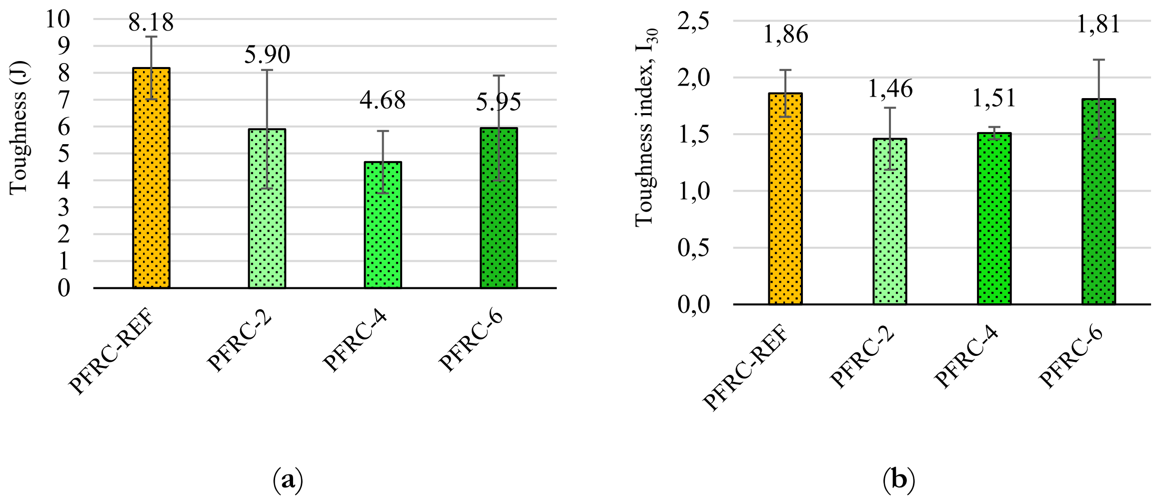

Figure 3.7. Values of toughness (**a**) and toughness index (**b**).

3.5. Conclusions

This study addressed the use of fibre from multilayer recycled plastic from food packaging plastic waste for reinforcement in concrete. Considering the results, the following conclusions can be drawn from this research:

- The physical properties studied were not significantly affected by the addition of recycled plastic fibres;
- The addition of RPF produces a certain decrease in compressive and flexural strength, but an improvement in the post-cracking properties of the concrete was achieved. The presence of plastic fibres inside the matrix increased both the ability of the concrete to resist the advancement of cracks, and its ability to deform plastically before breaking, giving the concrete matrix a certain residual tensile strength during the post-crack phase;
- The increase in toughness index with an increase in the percentage of recycled plastic fibres confirmed their stitching action inside the cement matrix, which is fundamentally dependent on the type of fibre and its quantity;
- The toughness index presented by the concrete with commercial fibres was greater than that with the same dosage of RPF, due to its rougher surface and greater adherence to the cementitious matrix. Furthermore, it should be noted that the toughness index relative to the mixture with 6 kg of recycled plastic fibres per cubic meter of concrete was comparable to that with commercial plastic fibre (PFRC-REF). This suggests that manufacturing RPF with a rougher surface could lead to a reduction in the amount needed to achieve similar toughness index levels to PFRC-REF.

In addition to the mechanical characteristics mentioned above, the use of recycled fibre from multilayer plastic waste derived from the packaging industry—which is currently difficult to recycle—in the production of this composite material could be an interesting approach to extend the life cycle of this material.

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A.B; project administration, J.A. and A.P.G.; funding acquisition, J.A. and A.P.G. All authors have read and agreed to the published version of the manuscript.

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4. Pollutant Potential of Reinforced Concrete Made with Recycled Plastic Fibres from Food Packaging Waste (Publication III)

Article

Pollutant Potential of Reinforced Concrete Made with Recycled Plastic Fibres from Food Packaging Waste

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Abstract

In our modern, fast-paced life, plastic is a versatile material essential to our economy; daily life is unthinkable without it. However, there are serious downsides for the environment and health, which are becoming more and more stark in our society, and the recycling of plastic offers a partial solution to these widespread problems. The present work delves into the environmental assessment of fibre-reinforced concrete specimens, made with recycled plastic fibres from food packaging waste. Leaching tank tests for the evaluation of the long-term release of pollutant elements, identification of leaching mechanisms, and the diffusion process of contaminants into the environment were conducted on fibre reinforced concrete. The results showed that the incorporation of the recycled plastic fibres, classified as non-hazardous, did not release relevant levels of any potential harmful element incorporated in concrete. Moreover, low mobility was detected in the studied elements and different release mechanisms were identified through long-term diffusion leaching tests. Hence, the environmental feasibility of the incorporation of recycled plastic fibre in concrete was proven. This study strengthens the objectives set out by the Circular Economy Action Plan, which includes the European Strategy for Plastics and aims to, among other things, boost the market for recycled plastics.

Keywords: sustainable construction; leaching tests; recycled plastic; reinforced concrete; circular economy

4.1. Introduction

Currently, facing the challenge of reducing the environmental impact resulting from our economic and social activities is a priority at all levels. The waste that is generated daily across the world is known. This waste has been studied in depth and new alternatives for recycling have granted some of its parts a second life cycle, e.g., recycled aggregates from construction and demolition waste, bottom ash from biomass combustion, or ash from solid urban waste. All of these examples refer to recycled materials that can be applied to engineering infrastructure. In addition, another example could be the utilisation of waste products for the manufacturing of recycled concrete: waste products, such as discarded tires, glass, steel, burnt foundry sand, and coal combustion by-products, have provided specific effects on the properties of fresh and hardened concrete [1].

However, there are numerous types of waste that are being generated in high quantities but, from an environmental point of view, they are not reused at acceptable rates. This is the case for plastic wastes coming from plastic food packaging, which is the subject of the present study. Plastic packaging is widely used in the food sector due to several properties: it is flexible and adaptable in form, hygienic, light, cost-effective, high versatility, and does not easily degrade [2]. Increasing its recycling potential is a priority.

In recent years, the circular economy of plastic has been increasing year by year and, in the specific case of plastic recycling, for the first time in Spain (in 2017), the number of tons of recycled plastic exceeded that of plastic deposited into landfills. These data demonstrate the increase in society's awareness, as well as the efforts and improvements of the collection systems, the modernisation of recycling processes, and the innovation of the plastics industry, which is incorporating more recycled material into new products. The plastics recycling sector in Spain is one of the most mature and powerful in Europe, with more than 120 companies and between 3500 and 4000 direct jobs, currently. The recycling of plastics will continue to grow, and it is estimated that, in 2030, with the recycling of 55% of plastic containers in Europe, savings of 14.8 million tons of CO_2 emissions will be achieved [3].

On the other hand, the construction sector is continuously evolving and reinforced concrete is an example of an innovative solution. Compared to other building materials, such as metal and polymers, concrete is significantly more brittle and exhibits poor tensile strength [4]. To overcome this deficiency, fibres have been added to concrete in recent decades [5,6], increasing the productivity and adapting more rational production techniques [7]. Fibre reinforced concrete (FRC) is composite material containing fibres in a cement matrix, either in an orderly manner or a randomly-distributed manner. Its properties are largely dependent on the type of fibre, fibre geometry, fibre content, orientation and distribution of the fibres, mixing and compaction techniques of the concrete, and size and shape of the aggregate [4]. The most common applications are pavements (slabs, industrial floors, or roadways), precast elements, and structural reinforcements. Different types of fibres have been used extensively in FRC (steel, polypropylene, glass, asbestos, polyester, etc.) [8], although steel and polypropylene are the most commonly used ones [9–12]. Polypropylene fibres (PF) are generally used in FRC structures to en- hance shrinkage cracking resistance, impact strength, spalling resistance, splitting tensile strength, and other properties [8,12–15]. Alhozaimy et al. [16] observed that an additive amount of 0.1% PF in concrete had a 44% increase in its flexural toughness. Mindness and Vondran [17] reported that compressive strength increased by about 25% at 0.5% volume fraction of PF in the concrete mixture design. Hughes and Fattuhi [18] suggested that compressive strength decreases but flexural properties are improved with increasing fibre content. According to Kakooei et al. [8], polypropylene fibres have hydrophobic levels, which protect them against wetting with cement paste, although their hydrophobic nature has no effect on the amount of water needed for concrete. The studies demonstrated that concrete made from macro plastic fibres is a composite material that could replace a part of the steel reinforcement in upstream concrete. Therefore, the exploration of the possibilities of reusing different types of plastic waste in concrete manufacturing would lead to unbeatable results while plastics are consumed daily, worldwide and the production of this waste has increased immensely during the past 50 years.

Nowadays, industrial by-products are widely used due to their ability to be recycled and valorised as secondary materials, which allows for the simultaneous saving of natural resources and energy [19]. Industries are finding new ways to use materials that would otherwise be discarded. Researchers have already used different recycled fibres to improve the behaviour of ordinary concrete. Ahmadi et al. [20] concluded that adding recycled steel fibres into concrete is feasible and recycled aggregates can be used in the production of structural concrete with a 50% replacement of aggregates. Gu and Ozbakkaloglu [21] evaluated the use of recycled plastic materials in conventional cement mortar; this concrete has been researched extensively. Plastics have been used in fibre-reinforced concrete in two forms: plastic aggregates (PA) and plastic fibres (PF). The use of recycled fibres in concrete can lead to improved concrete properties, in addition to increased recycling rates. Apart from PET, most types of common plastic (e.g., PP, PVC) are stable for decades, even for centuries, inside concrete. Review findings indicate that the use of recycled plastic mate- rials in concrete can contribute significantly to a more sustainable construction industry, but future studies on environmental aspects, such as the long-term behaviour of plastic materials in concrete and the environmental consequences of recycling of concrete containing plastics, is recommended. This is the focus of the present work. The incorporation of alternative materials in the production of concrete is liable to increase the quantity of pollutant elements in the product [22]. This fact could provoke an environmental risk due to the continued exposure of contaminated concrete to its surroundings [23–25]. Cement is a building material that has been extensively tested, with a long history of use under different environmental conditions; advantageous solidification results in a monolithic, watertight final product [26]. So, the use of potentially polluting materials in the manufacture of concrete would reduce the release of these in the case of contact with water; however, it could induce a loss of durability in the concrete [27].

Leaching tests have proved to be a useful tool for the environmental assessment of construction materials. Leaching is a diffusion–reaction phenomenon, which takes place when concrete is exposed to poorly mineralised or acid water [22,28]. Degradation consists of dissolution of calcium and hydroxide ions out of the matrix, which causes an increase in porosity and transport properties of surface concrete. Leaching and external sulphate attack on concrete leads to the dissolution of hydration products, mainly portlandite, and in cases of ingress of sulphate ions, to the formation of expansive products, such as gypsum and ettringite [27]. This emphasises the need to consider leaching behaviour in the design of concrete structures. Previous researchers in this area focused on the assessment of the underlying mechanisms of contaminant release and the chemical and physical factors that control leaching behaviour from construction materials under a wide range of environmental conditions [25,29,30]. Comparison with generally accepted norms, such as the European Council Decision 2003/33/EC [31], permits the assessment of whether application of a recycled product will be innocuous to the environment, although the release of substances from a material depends on their solubility, which is influenced by numerous factors including material composition, pH, permeability, contact time, and water/solid ratio [32–34].

With this framework, the present study aims to make a practical contribution to environmental sustainability and the circular economy, as it is focused on concrete made with non-commercial plastic fibres for reinforcement. Specimens of reinforced concrete were prepared using the plastic sheets used for food packaging at different weights per m^3 of concrete and two control specimens were used. In a preliminary laboratory phase, the mechanical performance of concrete made with the addition of recycled macro plastic fibres was evaluated and it was demonstrated that a certain improvement in the post-cracking properties of the concrete was achieved. The presence of plastic fibres inside the matrix increases both the ability of the concrete to resist the advancement of cracks and the ability to deform plastically before breaking, giving a certain residual tensile strength to the concrete matrix during the post-cracking phase. Once the mechanical assessment of using recycled plastic fibre as reinforcement in concrete was evaluated, the present study was implemented for the environmental assessment of RFC by means of leaching tank tests for the evaluation of the long-term identification of leaching mechanisms and the diffusion processes of contaminants into the environment. The following tests were performed: (1) compliance leaching test of recycled plastic fibres from food packaging waste for their pollutant potential classification, according to the EU Landfill Directive; (2) diffusion leaching test in tank for basic characterisation of pollutant release from concrete made with plastic fibres; and (3) dynamic diffusion leaching test for long term characterisation of pollutant release from concrete made with plastic fibres. All of these tests focused on the use of recycled materials coming from the processing of waste plastic used for packaging which, as highlighted by the Plastics Europe report, accounts for 45.4% of the packaging destined for recycling in Spain.

4.2. Materials

4.2.1. Recycled Plastic Fibres (RPF) from Food Packaging Waste (FPW)

The FPW consisted of macro plastic fibres obtained from the waste products and processing of plastic, supplied in waste rolls by a local company that deals with the production of plastic for packaging. The fibres used in the experiments were obtained in the laboratory by cutting them from plastic sheets. The average dimensions of the fibres were 50 mm long, 4 mm wide, and 0.125 mm thick (Figure 4.1).

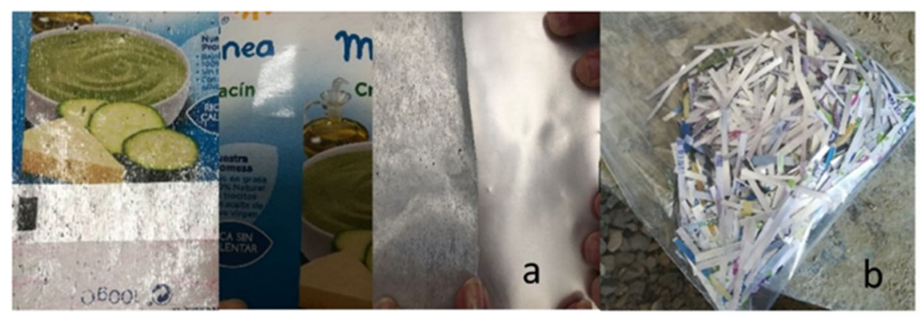


Figure 4.1. Plastic sheets from food packaging: front and back sides (**a**) and the prepared fibres (**b**).

The tested plastic sheets were made of four overlapping layers of different materials: polyester, aluminium, bi-axially oriented polyamide that is resistant to thermal sterilisation treatment (121 °C for 30 min), and polypropylene (PET + ALU + OPA STE + PP G). This is one of the most resistant multilayer complexes used in food packaging in terms of aluminium barriers. It is used for standing or flat bag formation, and also as tray lids. Aluminium makes it easily deformable; however, it does not lose the barrier effect. This composite material presents a high barrier to oxygen, humidity, aroma, and light, being rigid and opaque, sterilisable, and pasteurisable. The main properties and methods presented by the plastic sheet are summarised in Table 4.1.

Table 4.1. Technical data of plastic sheets PET + ALU + OPA STE + PP G.

Technical Data	Value	Unit	Method
Thickness	125.00 ± 10%	μm	ASTM E 252
Density	1.13 ± 10%	g/cm^3	ASTM D 1505
Tensile strength	500	MPa	ASTM D 882
Modulus of elasticity	9	GPa	ASTM D 882
Unit weight	140.90 ± 10%	g/m^2	ASTM D 252
Oxygen permeability	<0.5	$cm^3/m^2/24$ h	ASTM D 3985
Water vapor permeability	<0.5	$g/m^2/24$ h	ASTM F 1249
Permeability to carbon dioxide	<0.5	$cm^3/m^2/24$ h	ASTM D 3985
Permeability to nitrogen	<0.5	$cm^3/m^2/24$ h	ASTM D 3985
Departure sealing temperature	150-160	°C	-
Temperature for use	-18-121	°C	-

Footnote: ASTM: American Society for Testing and Materials.

4.2.2. Fibre Reinforced Concrete Specimens (FRC) Made with RPF

Specimens of FRC were prepared using the plastic sheets used for food packaging by the packaging company (Figure 4.2). This food packaging waste (FPW) was used at 2, 4, and 6 kg per m^3 of concrete, and the prepared mixtures were named using the codes: FRC2, FRC4 and FRC6. Two control specimens were also prepared: a control specimen of concrete without reinforcement (C-REF) and a fibre-reinforced concrete made with commercial fibres of polypropylene at a dosage of 2 kg of fibre per m^3 of concrete (FRC2-REF).

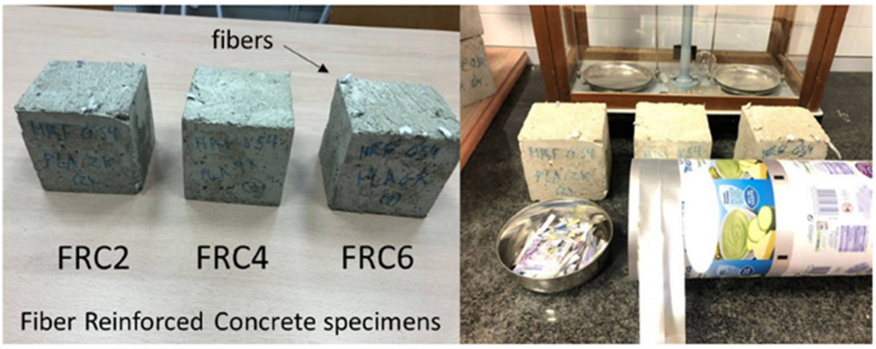


Figure 4.2. Concrete specimens of FRC with RPF: FRC2, FRC4 and FRC6.

The aggregates used for manufacturing the concrete mixes in the laboratory were coarse gravel (6–20 mm) and sand (0–4 mm), both of a siliceous nature. The cement used was CEM II 42.5 AV-R at 280 kg per m^3 of concrete produced, reaching a water to cement ratio of 0.54. To improve the workability and to avoid the segregation of the mixture, two different types of additives were used: the plasticiser Complast MR260 at 5 mL/kg of cement and the superplasticiser Structuro 357 at 7.7 mL/kg of cement.

4.3. Experimental Methods for Environmental Assessment

The use of any waste in concrete manufacturing for a second cycle life implies, not only the technical feasibility of a material, but also its environmental risk assessment. Leaching tests are an essential tool as they allow analysis of the transport rate of a constituent (pollutant element) through a material, which depends on the physical factors of the concrete, such as its porosity and permeability. Galvín et al. [35] observed that physical factors, such as density, porosity, and absorption, are not as relevant to the release of metals as was expected, which confirms the relevant role of chemistry on the release of metals. It proves the difficulty of reproducing leaching phenomena and the complexity of analysing the complete processes that govern the pollutant release of the chemical species being studied. Testing can be classified as tests aimed at attaining equilibrium conditions at the end of the leaching experiment (generally based on batch-type leaching tests or with controlled pH) or tests aimed at the dynamic aspects of leaching (tests in which time is an important variable, such as diffusion tests for monolithic materials and column leaching tests for granular materials) [24].
The experimental procedure performed on the FRC specimens was carried out accord- ing to the three different phases of study observed in Figure 4.3.

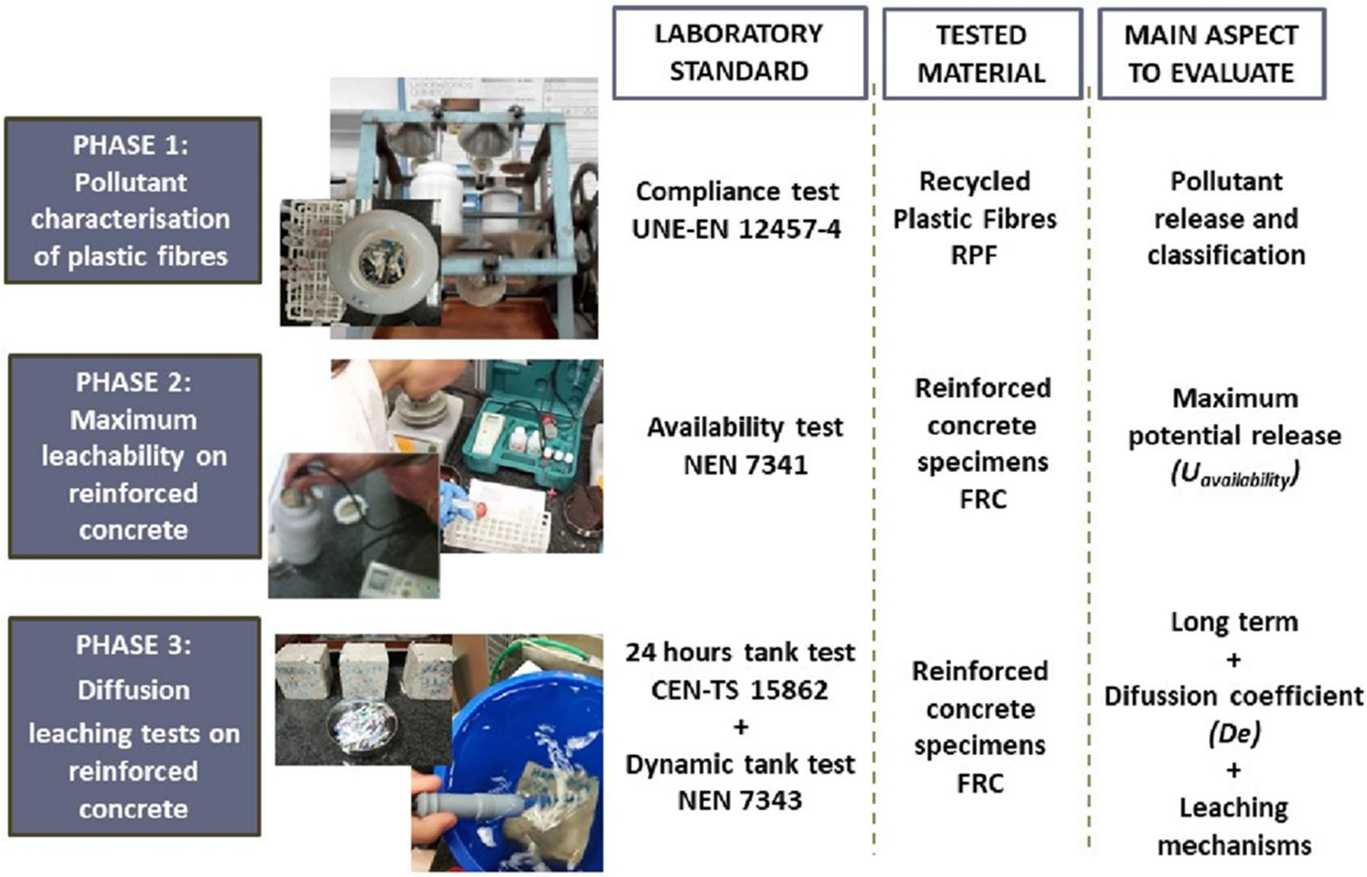


Figure 4.3. Phases of the experimental procedure for leaching assessment of FRC made with RPF.

- Phase 1: The Compliance Batch Test according to UNE-EN 12457-4:2003 for basic characterisation of leaching levels of a RPF sample. The data are compared with the legal limit established by Council Decision 2003/33/EC [31], which establishes limits for the release levels of 12 heavy metals (Ni, Cr, Sb, Se, Mn, Hg, As, Pb, Cd, Cu, Ba,and Zn) and three anions (sulphate, chloride, and fluoride), classifying the material as inert, non-hazardous and hazardous according to its pollutant potential.

- Phase 2: The availability test NEN 7371:2004 was performed in order to determine the maximum pollutant release from the tested materials. It was performed on ground samples of FRC2, FRC4, FRC6, C-REF, and FRC2-REF analysing three eluates per samples (at pH 4, pH 7, and the total, according to the standard).
- Phase 3: The standards conducted on monolithic samples (FRC2, FRC4, and FRC6) and the controls (C-REF, and FRC2-REF) were: the diffusion leaching tank test with periodic leachant renewal (UNE-EN 15863:2015) obtaining eight eluates per samples (40 samples in total) and the single stage batch leaching test (CEN-TS 15862:2012) analysing in ICP-MS one eluate per sample (five samples in total).

4.3.1. Compliance Test for RPF UNE-EN 12457-4:2003

The compliance test is a procedure for the basic characterisation of materials. The procedure was conducted on the RPF material by means of a single-step batch leaching test, in which the solution was shaken for 24 0.5 h at an L/S ratio of 10 L/kg. After the contact phase, the samples were left to decant, then filtered and a subsample of 40 mL of eluate collected for testing.

4.3.2. Determination of the Availability Threshold of FRC

The Dutch leaching test NEN 7371 was carried out on samples from FRC specimens after being crushed until they passed through a 125 µm sieve. The protocol consists of extracting the leachate in two steps with a liquid to solid ratio (L/S) of 50 L/kg each, at pH of 7 (first extraction) and pH of 4 (second extraction) and a contact time of three hour at each step. pH was kept constant by feedback control, adding 1 N HNO_3.

The purpose of the availability test was to indicate the quantity of a particular component that may leach out from a granular waste material exposed to extreme conditions (for example, in the very long term, after disintegration of the material, full oxidation and/or loss of acid neutralising capacity), in an aerobic environment. The potential risk depends on the availability of the contaminants for leaching [36,37].

The aim of this procedure is to establish pseudo-equilibrium conditions (eluent is in constant contact with the subsample of the granular material). The equilibrium is controlled by the solubility of the minerals present in the solid phase (once equilibrium is established, release is dependent on the geochemistry of the solid phase and the chemistry of the liquid phase, rather than on contact time.

4.3.3. Determination of the Leaching Behaviour of Monolithic Specimens

Leaching tests on concrete monoliths were conducted according to the 24 h tank test (CEN-TS 15862:2012), for basic characterisation of concrete specimens, and the dynamic Dutch standard with periodic leachant renewal (NEN 7343:2004), for long-term leaching assessment of concrete pieces.

In the NEN 7343:2004 standard, each monolith was placed in a plastic container and a given volume of de-ionized water was introduced to completely submerge the monolith and reach a "liquid to contact area ratio" (L/A ratio) of 8 cm^3/cm^2. The top surface of the monolith was kept at least 2 cm below the surface of the water and the distance between the surfaces of the monolith and the walls of the reactor was kept above 2 cm. At time intervals of 0.08, 1.00, 2.25, 8.00, 14.00, 15.00, 28.00 and 36.00 days, the aqueous solution was completely removed from the reactor and replaced with the same volume of deionised water. Before water renewal, the pH and electrical conductivity were measured, and a sample was extracted for testing. In the CEN-TS 15862:2012 standard, monoliths samples were contained in deionised water for 24 h with a L/A ratio of 12 cm^3/cm^2.

All of the samples that were collected from the different tests conducted were filtered through a cellulose–acetate membrane of 0.45 μm pore size and the solution was analysed within 24 h, for several elements and/or anions using ICP-MS and ionic chromatography, respectively.

In the diffusion leaching tank test in monolithic materials, release levels are expressed as leached quantity per unit area: E_i^*. defined as the measured leaching of a component in the fraction "*i*" (expressed in mg/m^2). This value can be calculated for each component

using Equation (1):

$$E_i^* = \frac{c_i \times V}{f \times A} \tag{1}$$

where:

- c_i is the concentration of the component in fraction *i* in μg/L;
- V is the volume of the eluate in L;
- A is the surface area of the test piece in m^2;
- f is a conversion factor: 1000 μg/mg.

Equation (2) allows the calculation of the cumulative values. The parameter ε_n^*, expressed in mg/m^2, measures the cumulative leaching of a component for a period (n) which comprises fraction *i* = 1 to *n*, *n* being the number of periods equal to the number of specified replenishment times (in this case, *n* = 8).

$$\varepsilon_n^* = \sum_{i=1}^{n} E_i^* \tag{2}$$

One of the main objectives of monolithic leaching testing is the determination of the diffusion coefficient, De, which can only be calculated when leaching is diffusion controlled. De, calculated according to Equation (3), is the effective diffusion coefficient which is used to assess the long-term leaching behaviour for an element in the monolith. For this calculation, it is necessary to use the availability data *($U_{availability}$)* estimated in Section 3.2.

$$D_e = \left(\frac{\varepsilon_{64}}{2653 \times \rho \times U_{availability}}\right)^2 \times f \tag{3}$$

where:

- D_e is the average, effective diffusion coefficient for a given component (m^2/s);
- ε_{64} is the derived cumulative leaching of the component over 64 days determined with Formula (4) (mg/m^2);
- ρ is the density of the test piece (kg dry matter per m^3);
- $U_{availability}$ is the leachable, available quantity derived according to NEN 7371 (mg per kg dry matter);
- f is a factor equal to 1.

We also express the average value of the effective diffusion coefficient in the form of a negative logarithm:

$$pD_e = -\log D_e$$

Equation (3), for the calculation of D_e coefficients, is in accordance with the leaching process described by Fick's law if (i) dissolution is extremely rapid compared to diffusion, (ii) leachant imbibition attains a fixed equilibrium value very soon after commencement of the elution test, and (iii) the embedded salt is fully soluble in the leachant. Given these preconditions, the following equation can be applied for the calculation of ε_{64}:

$$\varepsilon_{64} = \sqrt{64}\left\{\prod_{i=a}^{b} \frac{E_i^*}{\sqrt{t_i}-\sqrt{t_{i-1}}}\right\}^{\frac{1}{1+b-a}} \quad (4)$$

- ε_{64} is the derived cumulative leaching for a component over 64 days (mg/m^2);
- E_i^* is the measured leaching of the component in fraction i (mg/m^2);
- t_i is the end time of fraction i for which diffusion has been established, measured from the start of the test, in days;
- t_{i-1} is the start time of fraction i for which diffusion has been established, measured from the start of the test, in days;
- *a,b* are dimensionless indices by which an increment a-b is indicated for which a diffusion mechanism is established.

4.4. Results and Discussion

4.4.1. Release Levels on Leachates from RPF by UNE-EN 12457-4:2003

The release levels of pollutant elements were measured in the recycled plastic fibres (RPF) by performing the compliance test according to the UNE-EN 12457-4:2003 standard and the obtained data are shown in Table 4.2. Anions (sulphate, chloride, and fluoride) and heavy metals (Hg, Pb, Cd, and Se) are not included because their levels were lower than the detection limits.

Table 4.2. Leachate concentrations (mg/kg m.s.) from RPF by UNE-EN 12457-4:2003.

Leachate Concentrations (mg/kg m.s.) According to the Compliance Test

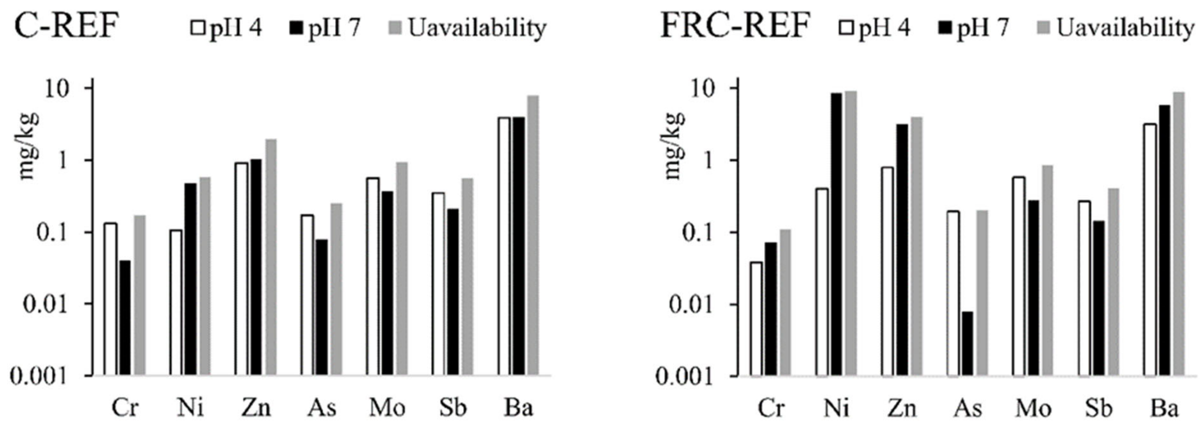
Elements	Inert Legal Limit at L/S of 10 L/kg	RPF
pH value	-	9.2
Cr	0.50	0.0035
Cu	2.00	0.0557
Zn	4.00	0.0528
Sb	0.06	0.0860
Ni	0.40	0.0074
Ba	20.00	0.1102
As	0.50	0.0088
Mo	0.50	0.0053

Low concentrations were measured for heavy metals and anions regulated by Council Decision 2003/33/EC [31], according to the compliance test, except for antimony. This surpassed the limit established for being classified as inert, and so FRP was classified as being a non-hazardous material. The release of antimony from PET is because it is widely used as a polycondensation catalyst in production [38].

4.4.2. Determination of Maximum Leachability for Heavy Metals by NEN 7371

Previous studies have demonstrated that the potential risk of environmental contamination from leaching is determined not by the total content of pollutants present in the material, but by the amount of water that can dissolve into the soil and reach the surface water and/or subsurface water [29,35]. Leaching tests are an essential tool for proving that the recycled material is safe and suitable for reuse.

The batch leaching tests in the UNE-EN 12457-4 standard allow measurement of the concentration of the polluting elements in the leachate, obtained by the laboratory procedures that simulate the effect of rainwater, surface water and groundwater coming into contact with the material used in the infrastructure [39]. The available leaching tests in NEN 7371 detects the maximum leachability of contaminants from the recycled concrete in the worst-case scenario, compared to the real situation of the tested material in the infrastructure. The leachable available quantity, $U_{availability}$, is then expressed in mg per kg dry matter, and was obtained and is represented as a logarithmic scale in Figure 4.4.



(a) **(b)**

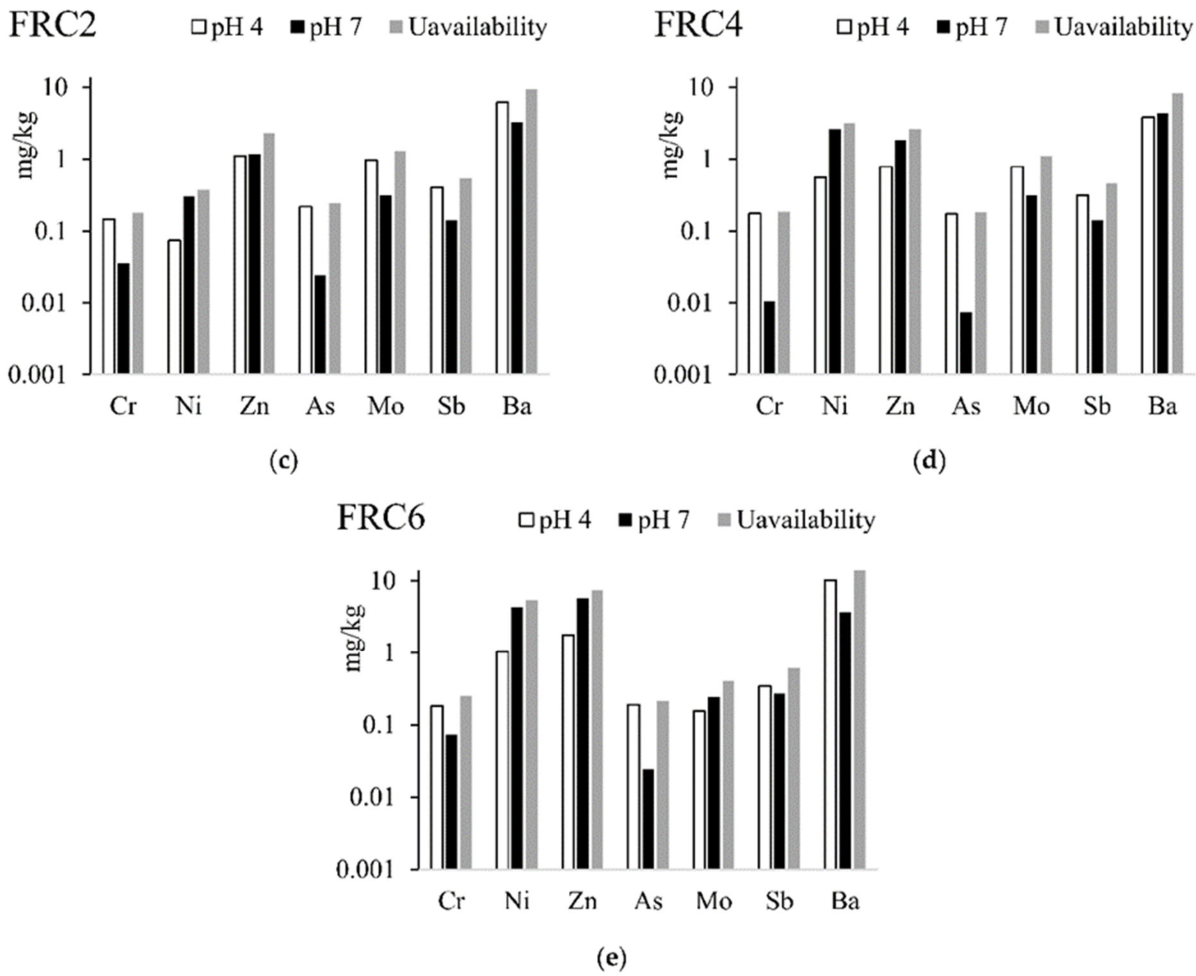


Figure 4.4. Comparison of two pH regimes in the availability test (pH 7 and pH 4) for heavy metals in samples: (**a**) control C-REF; (**b**) control FRC-REF; (**c**) FRC2 specimen; (**d**) FRC4 specimen; and (**e**) FRC6 specimen.

Regarding the effect of the percentage of RPF in the FRC specimens, the release levels observed for the elements studied were similar between the control specimens (C-REF without fibres and FRC-REF with a commercial fibre) and specimens FRC2 and FRC4. However, a slightly higher release was observed in the FRC6 samples for Zn and Ni.

After comparing the differences between the leachable quantity at pH 4 and pH 7, the strong influence of pH on release was confirmed, the pH in the environment being crucial in determining the release of many constituents in the monoliths and granular materials. As each material presents its own pH-dependent release curve, the leachability varies depending on the stage of the availability test and, according to Figure 4.4, elements such as Cr, As, Mo, and Sb present a higher release at pH 4, while heavy metals (such as Ni and Zn) showed higher release levels at pH 7. This is consistent with previous studies such as that of Tosti et al. [40]. Van der Sloot and Dijkstra [32] presented release curves as a function of pH which were similar and systematic for different groups of elements. Cations, such as Ni, Cu, Zn, Cd, Pb, Al, or Fe, present a release curve decreasing to pH 9 (with a minimum at this value) and increasing to pH 12. Anions, such as Mo, Cr, As, Se or Sb, present a curve with a minimum at pH 3, which increases to pH 10 and then flattens to pH 12. Finally, soluble salts have a distinct leach pattern, caused by their chemical speciation, and orders of magnitude vary as a function of pH. The results obtained are also consistent with Engelsen et al. [30], who observed that, for cations in the most acidic region (i.e., pH ~ 1–2), the leached contents for all elements are at their maxima and represent the available contents (maximum leachability) as the paste was degraded.

4.4.3. Diffusion Leaching Tests in FRC Specimens Made with RPF

4.4.3.1. Basic Characterisation: 24 h Tank Leaching Test by CEN/TS 15862: 2012

Leaching of monolithic materials is essentially governed by diffusion phenomena. Thus, for a previously basic characterisation, the diffusion leaching tank test was performed according to the CEN/TS 15862:2012 standard. This is a fast test (24 h) that allows the measurement of concentrations of the legally regulated elements, as shown in Table 4.3. Anions (sulphate, chloride, and fluoride) and heavy metals (Hg, Pb, Cd, Mo, As, Ni, and Se) are not represented because they are under the detection limits.

Table 4.3. Leachate concentrations (mg/m^2) from concrete specimens according to the diffusion leaching test.

	Leachate Concentrations from Concrete (mg/m^2)				
Elements	C-REF	FRC2-REF	FRC2	FRC4	FRC6
pH values	10.2	10.1	10.4	10.3	10.2
Cr	0.0042	0.0072	0.0030	0.0027	0.0032
Zn	0.0568	0.0495	0.0508	0.0576	0.0545
Sb	0.0149	0.0033	0.0105	0.0076	0.0059
Ba	0.0159	0.0058	0.0100	0.0061	0.0080

The 24-h dynamic tank test was carried out and it provided a basic and quick characterisation of the levels released by the concrete specimens. The heavy metals listed in Table 4.3 were released at low rates for all specimens and non-relevant differences are observed between the control specimen without fibre (C-REF) and specimens prepared with fibre reinforced concrete. The low release levels detected on the concrete specimens are justified, since solidification and stabilisation (S/S) using cement as a binder is most often applied (worldwide) for controlling the environmental impact of residues; it involves a conversion of the residues into a monolithic or granular material, ensuring easy handling and transportation to landfill sites. It also immobilises toxic pollutants by physical encapsulation, chemical incorporation and/or adsorption [41,42].

The data showed that the level of the heavy metals presented in Table 4.3 were similar to the RPF except to the antimony ones, which were lower than in RPF and inert limit, classifying them as inert material [43].

4. 4.3.2. Long Term Behaviour: Dynamic Tank Leaching Test by UNE-EN 15863: 2015

For a complete characterisation of the leaching behaviour of specimens in the long term, the dynamic tank leaching test was performed according to UNE-EN 15863:2015, to obtain different eluates over time ("*i*" fractions). To continue with the study of long-term leaching behaviour, the anions were not included due to their despicable release levels observed in previous sections. In the present section, the regulatory limits indicated by the Soil Quality Decree (SQD) [44] are used as a reference because this regulation specifies criteria for the (re)use of mineral materials in construction applications: "shaped materials", and the limits are most accurate to apply to FRC specimens (it prescribes the rules for the use of "shaped" construction materials).

Table 4.4 shows the *pDe*-values (negative logarithm of effective diffusion coefficient) for the elements which showed the highest mobility levels on concrete samples: Cr, Zn, Sb and Ba, along with cumulative emission (ε_{64}) and the pH at the start and the end of the test. The pH of the eluates collected did not vary significantly over the course of the test, presenting values corresponding to a medium alkaline, as expected. Only the elements Cr, Zn, Sb and Ba are represented because they present the higher mobilities. The cumulative emission for the elements in Table 4 did not present any substantial differences between the samples. The results obtained were consistent with the data obtained by previous researchers, in terms of cumulative emission at 64 days [28,45] and diffusion coefficient [46] in concrete.

Table 4.4. Cumulative emission (ε_{64}), diffusion coefficient (*De*) and *pDe*.

	Parameters	C-REF	FRC2-	FRC2	FRC4	FRC6
	Initial pH	10.2	10.4	10.3	10.2	10.4
	Final pH	10.1	10.2	10.2	10.1	10.2
Cr	Cumulative emission. ε_{64} (mg/m^2)	0.125	0.189	0.135	0.185	0.168
	Diffusion coefficient. D_e (m^2/s)	$1.500{\cdot}10^{-14}$	$8.270{\cdot}10^{-14}$	$1.580{\cdot}10^{-14}$	$2.800{\cdot}10^{-14}$	$1240{\cdot}10^{-14}$
	pD_e	13.820	13.080	13.800	13.550	13.910
Zn	Cumulative emission. ε_{64} (mg/m^2)	2.271	2.778	2.504	2.020	2.207
	Diffusion coefficient. D_e (m^2/s)	$3.770{\cdot}10^{-14}$	$1.380{\cdot}10^{-14}$	$3.410{\cdot}10^{-14}$	$1.660{\cdot}10^{-14}$	$2.480{\cdot}10^{-14}$
	pD_e	13.420	13.860	13.470	13.780	14.610
Sb	Cumulative emission. ε_{64} (mg/m^2)	0.717	0.643	0.861	0.342	0.729
	Diffusion coefficient. D_e (m^2/s)	$4.580{\cdot}10^{-14}$	$6.940{\cdot}10^{-14}$	$7.010{\cdot}10^{-14}$	$1.600{\cdot}10^{-14}$	$3.970{\cdot}10^{-14}$
	pD_e	13.340	13.150	13.150	13.800	13.400
Ba	Cumulative emission. ε_{64} (mg/m^2)	4.261	3.965	3.992	3.277	3.403
	Diffusion coefficient. D_e (m^2/s)	$8.130{\cdot}10^{-14}$	$5.480{\cdot}10^{-15}$	$5.100{\cdot}10^{-15}$	$4.530{\cdot}10^{-15}$	$1.710{\cdot}10^{-15}$
	pD_e	14.090	14.260	14.290	14.340	14.770

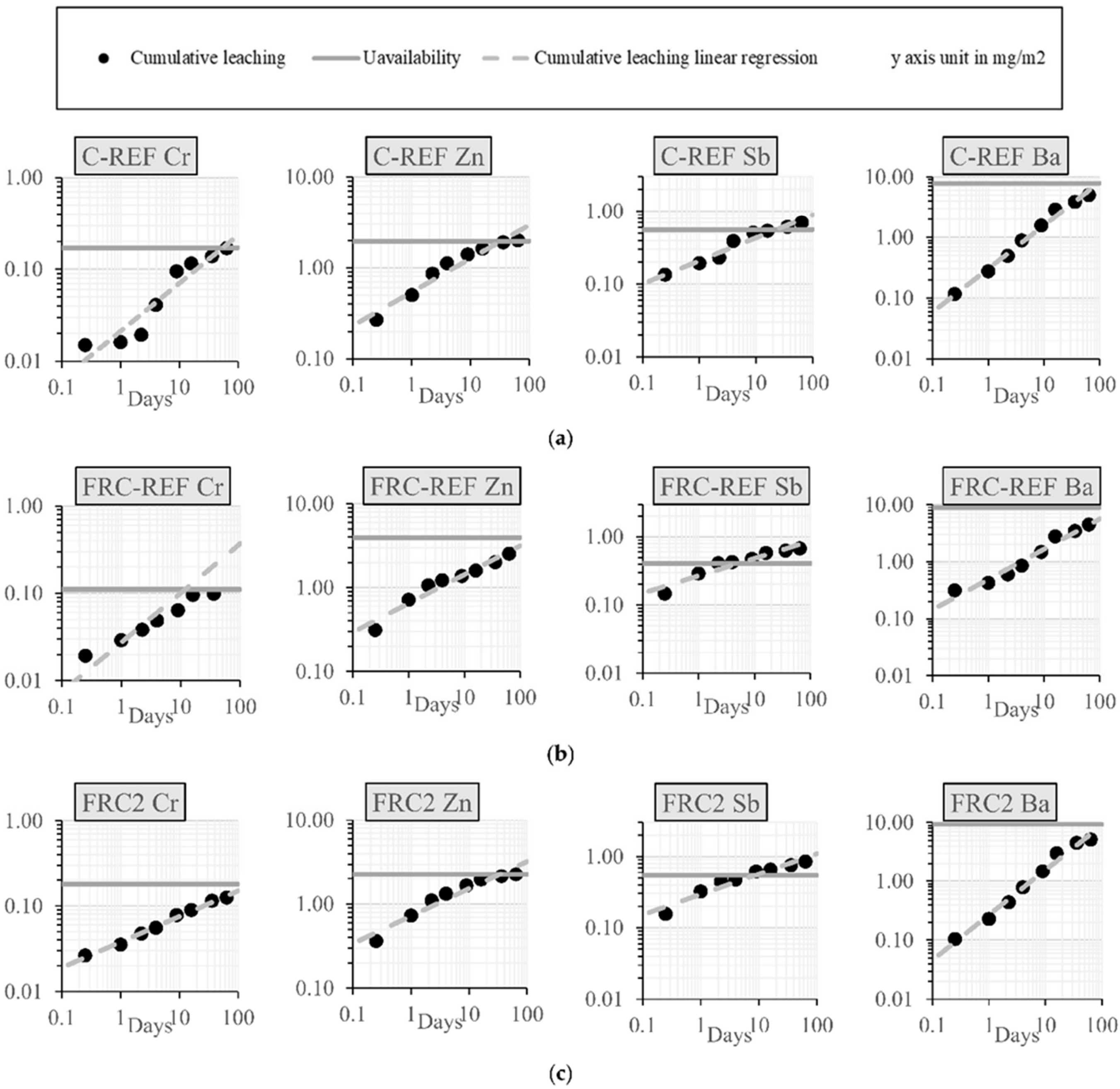
The higher pD_e value implies the lower speed of leaching of the component concerned with constant availability $U_{availability}$. The pD_e value determines the concentration gradient, which is the driving force for diffusion of the pollutant elements studied: $pD_e > 12.5$: com- ponent with low mobility; $11.0 < pD_e < 12.5$: component with average mobility; $pD_e < 11.0$: component with high mobility, according to the NEN 7343. Of all the represented elements: Cr, Zn, Se and Ba showed a pD_e value higher than 12.5 because they are components with low mobility.

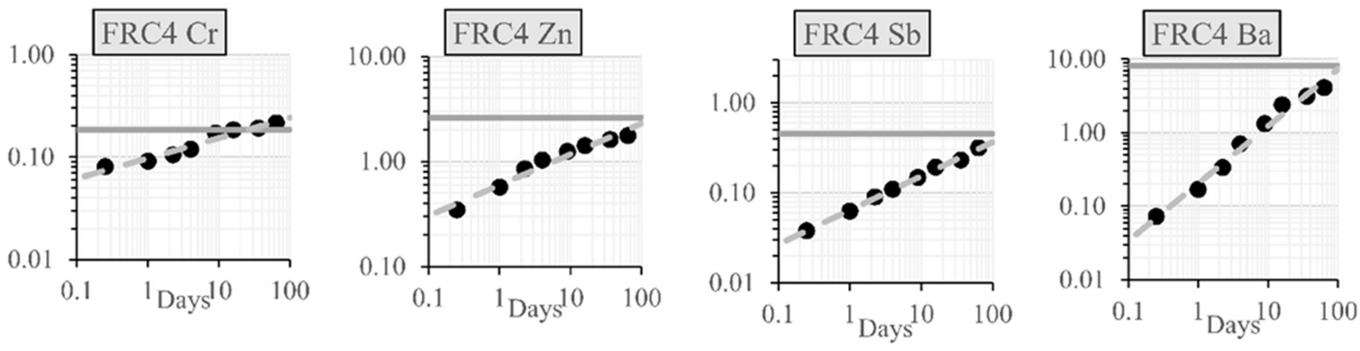
The determination of the leaching mechanisms occurring in the diffusion test is possible, based on the leaching of components previously measured and establishing whether the matrix of the test piece is dissolving during the conduct of the test. It is possible to determine whether leaching is diffusion controlled or whether other leaching mechanisms also contribute, for all individual components. Despite the usefulness of short- term tests, they do not provide information about the behaviour of contaminants in the medium or long-term. On the contrary, dynamic tests are usually employed for elucidating the dominant leaching mechanism from a stabilised/solidified (S/S) waste [47,48]. As a matter of fact, the main release mechanisms are: (i) solubility; (ii) diffusion from the internal porosity of the matrix to the surface; and (iii) surface wash-off (where substances concentrated at the surface of the monoliths may be released at the first contact with water) [49].

Figure 4.5 illustrates the graphical representation of diffusion-controlled leaching (mg/m^2) vs. time (days) for the identification of leaching mechanisms of the four heavy metals studied. The Uavailability value obtained by the availability test is included to observe the maximum release level obtained for each heavy metal and each specimen is plotted in Figure 4.5, along with the obtained data from the diffusion test of cumulative leaching measurements (expressed in mg/m^2) and its linear regression. Regarding the leaching behaviour of monolithic samples, there are three main basic transport and chemical mecha- nisms that usually control the leaching processes: surface wash off, where way the most soluble phases on the surface of the monolith are released quickly, diffusion transport of the solubilised species in the aqueous phase inside the monolith, and the surface dissolution of the monolith in contact with the aqueous phase that surrounds it (chemical mechanism control) [32,48].

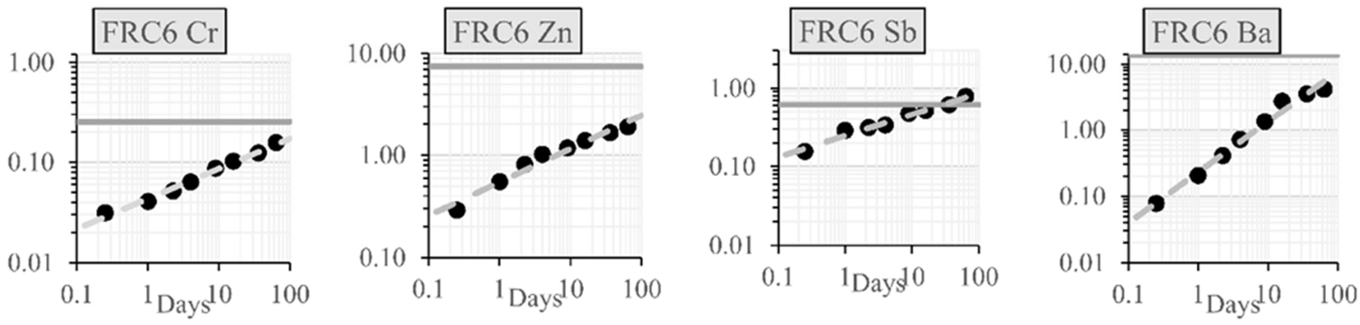
For determining the main leaching mechanism, the linear regression that fits the experimental points of cumulative fraction released versus time, or the logarithmic representation of cumulative flux versus time, can be used [50–53]. According to Torras et al. [48], if the slope of the straight line fitting the experimental data is greater than 0.65, then surface dissolution is the dominant leaching mechanism; if the *slope* is close to 0.50 (0.35 < *slope* ≤ 0.65) diffusion transport is the leaching mechanism and if the *slope* is near 0 (*slope* ≤ 0.35) a phenomenon of initial wash-off or depletion (if it happened in the middle or at the end of the test) is occurring.

According to the data obtained, all element concentrations in the leachates are be- low the permissible limits prescribed by the SQD and used as a reference for shaped construction materials, such as concrete. The limits for heavy metals are shown in Table 4.5.





(d)



(e)

Figure 4.5. Graphical representation of diffusion-controlled leaching (mg/m^2) vs. time (days) in samples: (**a**) control C-REF; (**b**) control FRC-REF; (**c**) FRC2 specimen; (**d**) FRC4 specimen; and (**e**) FRC6 specimen.

Table 4.5. Limits for diffusion release levels from shaped construction materials (in mg/m^2). Slope of linear regression andcorrelation coefficient.

	Parameters	C-REF	FRC2-REF	FRC2	FRC4	FRC6
Cr	SQD limit for shaped materials emission limits ε_{64} (mg/m^2)			120.0		
	Slope of the linear regression	0.522	0.566	0.299	0.197	0.299
	Correlation coefficient, R^2	0.907	0.739	0.992	0.951	0.989
	Standard deviation	0.146	0.294	0.023	0.039	0.027
Zn	SQD limit for shaped materials emission limits ε_{64} (mg/m^2)			800.0		
	Slope of the linear regression	0.369	0.343	0.326	0.293	0.327
	Correlation coefficient, R^2	0.944	0.946	0.936	0.960	0.963
	Standard deviation	0.078	0.072	0.075	0.052	0.056
Sb	SQD limit for shaped materials emission limits ε_{64} (mg/m^2)			8.7		
	Slope of the linear regression	0.317	0.255	0.287	0.378	0.272
	Correlation coefficient, R^2	0.951	0.911	0.936	0.997	0.977
	Standard deviation	0.063	0.070	0.075	0.017	0.036
Ba	SQD limit for shaped materials emission limits ε_{64} (mg/m^2)			1500.0		
	Slope of the linear regression	0.716	0.530	0.763	0.777	0.764
	Correlation coefficient, R^2	0.989	0.962	0.986	0.983	0.984
	Standard deviation	0.065	0.091	0.079	0.090	0.086

According to the data in Table 4.5, Zn and Sb showed linear regression slopes that lead to the idea that the wash off mechanism occurred. A flat Zn curve appeared in the last steps of the diffusion test, indicating that the element has been depleted in dissolution. Ba presented a tendency towards a dissolution mechanism, except in the FRC-REF sample. In Figure 4.5, the cumulative leaching points follow the slope and the release levels increase along the steps. The element Cr presented lower values for the samples with RPF, corresponding to the wash off mechanism, whereas diffusion was detected for control specimens as has been observed in previous works: contaminants may be in the oxidised or reduced form (e.g., Chromium may be present as CrO_4^{-2} or Cr^{+3}). Heavy metals tend to be complex and strongly associated with the natural, humic substances present in natural waters or soils and these complex forms of heavy metals are generally highly soluble, being released more rapidly than simple forms [32]. Thus, comparing Cr and Ba patterns, it is clearly observed that the slope is more horizontal for Cr (diffusion) than for Ba (dissolution), and the difference in the release pattern is clearly observed for these elements with the differentiated mechanisms.

4.5. Conclusions

Waste plastic food packaging, used for the preparation of plastic sheets to be used as fibre reinforcement in concrete, showed low release levels of the elements determined by the Council Decision 2003/33/EC and they were under the inert limits, except for antimony. Hence, they are classified as non-hazardous material. The environmental feasibility of RPF was proven when added to the manufacturing of reinforced concrete with macro recycled plastic fibres at different amounts (2, 4 and 6 kg per m^3). The low release levels in concrete with RPF confirms that the process of stabilisation/solidification (S/S) of the waste in the cement matrix of the concrete, reduces the concentrations in leachatesto a minimum. The concentrations of the elements with higher mobility, in the long-term, were much lower than the limits imposed by regulations for “shaped” construction materials, such as SQD.

The study carried out to identify the transport and chemical mechanisms, indicated that the release patterns were: wash-off for Sb, Zn (although Zn also presented depletion in the final steps) and Cr in concrete samples with RPF; dissolution for Ba, except for FRC-REF samples; and diffusion for Cr in FRC-REF and C-REF samples. These were identified in the curves of the cumulative leaching vs. time graphs, obtained by conducting the dynamic tank leaching test.

Therefore, the present study contributes to the recycling of wastes, such as plastic packaging with a thickness of 0.125 mm and layers of different polymers, as reinforcement in concrete. This type of material does not present any possible environmental impact and could be a viable alternative that would help increase the valorisation of such types of plastic residue, which is considered as being one of the biggest concerns worldwide.

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5. Main results and general discussion

This chapter presents the main results obtained in this Doctoral Thesis focused on finding a second life to wastes from multilayer food plastic packaging and at the same time to experiment with the use of innovative construction materials in the engineering and construction sector.

Previous researchers have been working on finding innovative and sustainable solutions to address the enormous quantities of plastic waste that are produced every year which, after being collected, are transformed into energy, recycled, or sent to landfills. Providing the possibility of recycling to plastic waste as a material to be incorporated, in the form of macro-fibres, into concrete, could be one such solution.

In recent years, the circular economy in the framework of 2030 Agenda has been applied to plastic wastes. Research works, which focused on reducing the production of this type of waste, has been increased last years. The plastics recycling sector in Spain is one of the most mature and powerful in Europe, and it is estimated that, in 2030, thanks to the recycling of most than a half of plastic containers in Europe, million tons of CO_2 emissions will be achieved.

For many decades, researchers have been working on finding innovative and sustainable solutions to address the enormous quantities of plastic waste that are produced every year. In that sense, the incorporation of plastic wastes as recycled fibres (in the form of macro-fibres) into concrete, could be one such solution as it is demonstrated in the present Doctoral Thesis. These plastic fibres are synthetic fibres that are known to help improve the ductility, crack resistance, and toughness of conventional concrete.

This thesis has contributed to prove the mechanical feasibility of using recycled plastic fibres in concrete in terms of toughness, bringing an improvement in the post-crack resistance of the composite material. On the other hand, it was proven that the incorporation of the recycled plastic fibres was viable from an environmental point of view based on leaching tests.

The experimental methodology performed was based on laboratory test for testing the physical and mechanical properties as: dry density and the open porosity, Compressive Strength, Elastic Modulus, Flexural Strength, CMOD and Toughness (Chapter 3). For the environmental evaluation, different leaching procedures were followed with the aim of reaching different levels of analysis: Compliance leaching Test for basic characterisation; the availability test to determine the maximum pollutant release and leaching tank test to study the release levels at long-term (Chapter 4).

Mechanical Performance of Concrete Made with the Addition of Recycled Macro Plastic Fibres (Publication II)

The aim of the research was to investigate the physical and mechanical characteristics of a concrete reinforced by adding, in the mixing phase, plastic macro fibres obtained from the multilayer plastic food packaging industry waste .

For this purpose, different batches of concrete specimens were produced in the laboratory of cubic (100 mm length), cylindrical (300 mm height, 150 mm diameter) and prismatic (400 mm length, 100 mm width, 100 mm height): (1) specimens without the addition of fibres (C-REF) and (2) specimens made with the addition of commercial macro plastic fibres (PFRC-REF), with a fibre dosage of 2 kg/m^3, used as reference mixes; and (3) specimens with recycled plastic macro fibres with increasing dosage of 2 kg/m^3 (PFRC-2), 4 kg/m^3 (PFRC-4) and 6 kg/m^3 (PFRC-6). The recycled plastic macro fibres (RPF) were mechanically obtained directly from multilayer films from a Spanish plastic food packaging manufacturing company. The dimensions of the fibres obtained were 50 mm in length, 4 mm in width and 0.125 mm in thickness and consist of four layers of overlapping materials: polyester, aluminum, biaxially oriented polyamide, and polypropylene (PET + ALU + OPA STE + PPG).

As regards the study of the physical characteristics of the concrete produced, dry density and the water absorption were taken into consideration.

The mechanical point of view examined the dosage of RPF that most characterize concrete for its use as a building material in the civil engineering sector: compressive strength, the elastic modulus, the flexural strength and toughness. The compression tests were carried out both on the cubic-shaped specimens with a curing of 7 and 28 days and on the cylindrical-shaped specimens with curing of 28 days. The determination of the elastic modulus was carried out on cylindrical specimens at 28 days of aging. The flexural strength and toughness were obtained by bending tests on four points on prismatic specimens with notch in the central lower part and with a curing of 28 days.

The results obtained from the aforementioned tests have shown that the physical characteristics of the concrete, analyzed in the present study, were not altered significantly by the presence of the RPF, deviating slightly from those of ordinary concrete.

As regards the mechanical characteristics, the tests have shown that, in the compression test, the strength of the concrete with the presence of RPF has undergone a decrease compared to the comparative concrete (C-REF) and that this decrease has been increasing with increasing RPF dosage. A greater drop in strength was found in the cylindrical specimens, probably caused by the different packaging methods compared to the cubic ones. The results relating to the modulus of elasticity have shown how the values obtained in the case of the PFRC are similar to those of the C-REF, a result that has also been obtained by other authors in previous research. The flexural strength of the PFRC denotes a considerable decrease, with the increase in the dosage of fibre, compared to the C-REF. This decline, as also claimed in other studies, could be due to the presence of voids within the concrete matrix. The toughness of the PFRC was obtained from the experimental diagrams obtained by measuring the crack mouth opening displacement (CMOD) at the point values of the force applied to the prismatic specimen carved in the central part. From the analysis of the data obtained, there was an increase in the resistance capacity of the PFRC during the post-cracking phase. This resulted in a consequent increase in the material's ability to resist the advancement of the formation of cracks and allowed the PFRC to absorb energy and deform plastically before the final failure of the specimen. Furthermore, as the RPF dosage increased, an increase in the toughness index (I_{30}) was seen compared to that of C-REF, the latter being set equal to one, denoting how the stitching action of the fibres inside the cementitious matrix depends on the type of fibre used and its quantity.

In conclusion, it can be stated that the present research has shown that the use of PFRC, as a building material for civil engineering works, could be a valid alternative to traditional concrete, especially regarding resistance during the post cracking phase during the which is felt the reinforcement due to the RPF. If to this advantage, it is added the fact that with the use of RPF, inside the concrete, it contributes to the recycling of a plastic material that is currently difficult to recycle.

Pollutant Potential of Reinforced Concrete Made with Recycled Plastic Fibres from Food Packaging Waste (Publication III)

The purpose of this work has been to contribute to finding a sustainable solution, from an environmental point of view, to the growing problem of recycling waste materials. Among these, a prominent place is occupied by the plastic food packaging sector which alone generates large quantities of waste which, if not recycled, should be disposed of in landfills, with consequent damage to both the environment and humans.

Plastic fibres, both micro and macro, are used as an addition to the concrete in the construction sector, with the function of reinforcing the material itself. The present research aims to evaluate the environmental impact of the use of recycled plastic macro fibres from plastic food packaging. With the premise that the plastic material used in this study is initially a material free of substances potentially dangerous for the environment, given that it is used in direct contact with substances and products intended for human consumption, the goal was to evaluate its potential harmfulness once used as a reinforcement within a solid matrix such as concrete for civil constructions. Concrete reinforced with plastic fibres (FRC) coming from plastic food packaging waste, in contact with atmospheric agents and with the ground itself, could release chemical substances which, flowing into the ground and in the aquifers.

The phases used in the experimentation concerned: (1) the evaluation of the release levels on leachate by recycled plastic fibres (RPF), (2) the determination of maximum leachability for heavy metals in the FRC according to the dosage of RPF and the pH of the environment in which the material will be used, (3) the diffusion leaching tank test for monolithic materials, (4) the long term behaviour of the specimens by the dynamic tank leaching test. All the experimental procedures were conducted following the standard laboratory of the Spanish, Dutch and European standards.

The conclusions to which the results of the previous laboratory phases led allow us to state that: (1) the leachate concentrations from RPF used in the concrete is for all the chemical elements evaluated (Cr, Cu, Zn, Sb, Ni, Ba, As, Mo) very low and in any case considerably lower than the limits established by Spanish and European legislation; (2) regarding the leachable available quantity (Uavailability) of the FRC specimens it is observed that the release levels measured are similar to those of the control specimens without fibre (C-REF) and to those of the FRC-REF with commercial fibres, with a slightly higher release for the FRC6 samples for Zn and Ni; in addition, the results obtained confirm the strong influence that the pH of the environment in which the FRC is located for the purpose of releasing many elements that make up the materials, both monolithic, as in our case, and granular ones; (3) the release of heavy metals (Cr, Zn, Sb, Ba) by the FRC specimens compared to both the comparative C-REF, without fibre, and to those of FRC2-REF, with commercial fibre, did not show significant differences, justifying this as the control of the environmental impact of waste can be implemented by transforming them into monolithic materials, as is the case of the FRC used in this research, or granular, thus ensuring the immobilization of any toxic elements by physical encapsulation, chemical incorporation or adsorption; (4) using the regulatory limits indicated by the Soil Quality Decree (SQD) for the "shaped" construction materials such as FRC specimens, the dynamic tank leaching test shows how the analysed elements (Cr, Zn, Sb, Ba) have demonstrated to have all a low mobility within the concrete being the effective diffusion coefficient (pDe) for all greater than the limit of 12.5, according to the NEN 7343. The results of the tests have also shown that, in the long-term behaviour, the main leaching mechanism of the analysed elements contained within the FRC, was the surface dissolution mechanism for the Ba, and the wash off mechanism for the Zn, Sb and Cr.

Based on the foregoing, it can be stated that the use of FRC using plastic food packaging as a reinforcing fibre does not cause any damage to the environment because the fibre material used is classified as a non-hazardous material and therefore safe and usable, both because the release of potentially harmful substances falls largely within the limits established by Spanish and European standards on environmental control. In this way, a residue that would otherwise be difficult to recycle is enhanced.

6. Conclusions

6.1. General conclusions

Based on the scope of this work and after developing the methodology described in Chapter 2, the following general conclusions are presented:

1. The literary review carried out on the use of recycled plastic fibres as reinforcement in concrete allowed stablishing the starting point of the present research work, since previous studies performed on recycled plastic macro fibres demonstrated that the data of physical parameters as workability or density and results of mechanical properties as elastic modulus, durability and compression and flexural tensile strength proved the feasibility of reuse plastic wastes as reinforcement plastic fibres for concrete manufacturing.
2. The mechanical performance of reinforced concrete with addition of recycled macro plastic fibres conducted in laboratory showed that although a certain decrease of compressive and flexural strength was obtained, an improvement in the post-cracking properties of the tested concrete was achieved. It was reported the effect of the addition of recycled fibres in the matrix, since when the percentage of recycled plastic fibres arose, an increase in toughness index were observed which confirmed their stitching action. Furthermore, despite of the fact the toughness index in recycled macro fibres were lower that the presented by the concrete prepared with commercial fibres (mainly due to its rougher surface), there was an increment as the percentage of recycled plastic fibre arose. It leads to the idea that manufacturing recycled plastic fibres with a rougher surface could lead to an increase in toughness; hence, a reduction in the amount needed to achieve similar toughness index levels to the concrete with commercial fibres.
3. The environmental study based on leaching tests demonstrated that the fibres prepared from the plastic wastes can be classified as non hazardous waste according to the legal limits of the Council Decision 2003/33/EC. Regarding the pollutant potential of the reinforced concrete, the diffusion leaching tests performed on the monoliths confirms that the incorporation of this waste into the cement matrix of the concrete reduces the concentrations in the leachates. Finally, the study of the leaching behaviour at the long term of the different concrete specimens allowed the analysis of the release patterns of the elements studied, identifying the three transport mechanisms of the heavy metals: wash-off, dissolution and diffusion phenomena.

Therefore, the present Doctoral Thesis within the framework of the economic, social and environmental challenges demanded by today's society, contributes to the recycling of plastic wastes, a global concern which requires a coordinated, international and urgent response involving all relevant actors at different levels.

6.2. Future lines of research proposed

The carrying out of the experimental methodology described above and the scientific productivity resulted from the findings of the present Doctoral Thesis contribute to the sustainability of the plastic and construction sector combining: (1) recycling a multilayer plastic wastes that would be discarded by the packaging company and (2) adapting these through cutting for preparing recycled plastic fibre that can be added in concrete to be used in reinforced concrete promoting their use by the construction sector producing reinforced concrete mixtures with recycled multilayer plastic fibres that not only are technical but also economic and environmental feasible in comparison to the conventional reinforced concrete.

Thus, the following future lines are proposed at the present Doctoral Thesis:

- To improve the recyclability potential of the multilayer plastic wastes from food packaging company. To design a full-scale prototype to be incorporated in the factory process for the industrialization (handling and preparation) of the food packaging wastes for its transformation into macro fibres of different sizes and shapes to ensure the potential use of this waste in reinforced concrete.
- To deep on the study and analysis of the effect of the dimensions of the macro fibres, dosage and adhesion between the fibre and the concrete matrix on the mechanical and physical properties of reinforced concrete with this type of macro fibres to obtain the optimal mixtures in laboratory redefining the traditional reinforced concrete mixtures.
- To adapt the mentioned characteristics of the recycled fibres to the real demand of the precast sector in order to manufacture the precast concrete pieces with optimal technical characteristics.
- Quantification of the economic and environmental benefits that providing a second life cycle to these wastes in relation to the use of conventional macro fibres. In this sense, the application of the Life Cycle Analysis (LCA) methodology to be able to establish the general comparison of a conventional reinforced concrete made with commercial fibres versus a concrete reinforced with recycled fibres is really interesting to quantify the environmental, social and economic benefits that the use of recycled macrofibres from food packaging waste produces in relation to the use of conventional ones.