

Innovative concretes for low-carbon constructions: a review

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Abstract

The article reviews the recent research contributions and future promising perspectives regarding innovation in concrete technologies for low-carbon applications in buildings. To this aim, an original classification of recent trends is presented for reducing the carbon footprint of concrete constructions by identifying three main research lines. The first one is related to the enhancement of physical and mechanical properties of concrete, the second one is related to resource efficiency and raw materials' saving and the third one concerns the role of smart concretes in building energy efficiency. Possible synergies between the three addressed main research lines are finally discussed.

Keywords: concrete; cement-based material; smart material; energy efficiency in buildings; sustainable building material

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1 INTRODUCTION

The environmental impact of the construction sector is enormous, which is mostly associated with consumption of natural materials, emission of greenhouse gases, primary energy consumption and waste production. Literature studies reported that the building industry contributes to 24% of the total material extractions worldwide [1]. Furthermore, it was reported that ~40% of the materials in the global market are consumed by activities related to the construction sector, and that the same activities heavily contribute to greenhouse gases emissions and to the formation of acid rains [2].

Concrete plays a major role in the construction industry, being the most widespread construction material worldwide used for buildings, infrastructural systems, geotechnical works, industrial plants, road pavements, water dams and more, whereby it was calculated that ~1 ton of concrete is produced every year for every person in the world [3]. The large use of concrete is motivated by its low cost, versatility of use and good mechanical properties when working in compression and used to withstand vertical loads. Owing to these features, the main weaknesses, shortcomings and limitations of such material and, primarily, its well-known

low tensile strength, have not significantly affected its use so far. This also explains why concrete industry is a key player in the global emissions of carbon dioxide, to which it is acknowledged to contribute by a percentage of ~7, whereby 90% of such emissions are related to cement fabrication [4]. Such a huge contribution of the construction industry to excessive releasing of greenhouse gases in the atmosphere is stimulating scientific researchers, as well as decision-makers, toward the development of innovative concretes with reduced environmental impact. In this regard, Müller *et al.* [5] observed that enhancing the sustainability potential of building materials, in general, and of concrete, in particular, is a multidisciplinary task that involves three main parameters that interact with each other, namely environmental impact, technical performance and lifetime. This is especially true for concrete and demonstrates that sustainable are those concretes that improve at least one of such parameters, without negatively affecting the others. The environmental impact and life-cycle assessment (LCA) of concrete, including its 'green' variants, was theoretically investigated by Van de Heede and Belie [6], showing that the final score can be significantly affected by the main assumptions made in the LCA calculations. It follows that the improvements in concrete technologies resulting in actual mitigations of the environmental

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impact of concrete constructions are quite challenging and require a multidisciplinary expertise to properly engineer the material in such a way to achieve impact mitigation through the whole lifespan of a structure. This article is contribution to this topic, by presenting a review of the most recent research results and scientific trends toward the reduction of the environmental impact of concrete constructions. To this aim, an original classification of sustainable concretes is proposed (Figure 1), which does not only consider aspects that are directly related to primary energy consumption and greenhouse gases emissions, but also considers other aspects such as mechanical performance and durability.

The rest of the article is organized as follows. Section 2 analyzes special concretes whose environmental impact is mitigated by enhancing their physical and mechanical properties. These include (i) high strength concretes (HSCs) that allow to reduce the overall structural volume to withstand the loads, (ii) concretes with enhanced durability, (ii) lightweight concretes (LWCs) and other interesting developments. Section 3 discusses the main research trends devoted to enhancing resource efficiency of concretes, including recycling of raw materials, mitigating the environmental impact related to the production of cement, reuse of industrial waste and more. Section 4 presents an overview on special concretes for energy efficiency, including concretes with optimized thermal behavior, concretes with improved optical properties for passive cooling applications and smart concretes for energy harvesting, just to mention a few of them. Finally, a discussion about the main possible synergies among the presented innovative contributions is presented, by highlighting potentialities for further research progress of different technologies and their market perspectives.

2 CONCRETES WITH ENHANCED PHYSICAL AND MECHANICAL PROPERTIES

2.1 High strength concretes

The use of concretes with high mechanical strength leads to more sustainable constructions with increased long-time

serviceability. HSC allows to reduce the cross-sections of the structural members and consequently the overall volume of the material required for the whole building [7]. The new family of ultra high performance concretes (UHPCs) is reaching a compressive strength similar to that of steel, allowing the design of slender structures [8, 9]. However, their mix design needs to be accurately calibrated in order to reduce high-energy-intensive components. Harbert and Russel [10] estimated the material reduction volume achieved, thanks to the enhancement of the concrete mechanical strength, in order to evaluate the corresponding reduction of the CO₂ emissions amount.

Nevertheless, the increase of mechanical strength of concrete is associated with the use of a greater quantity of typically high emission materials. Indeed, mechanical strength is about proportional to the square of the cement content, kept constant the other variables, and therefore it results proportional to the square of CO₂ emissions. Due to the decrease of the amount of concrete needed to manufacture a given structural component, a 30% reduction of emissions when doubling normal concrete compressive strength can be estimated, while the use of UHPCs determines a reduction of 50% of the same CO₂ emissions [10]. Van der Heede *et al.* [6] evaluated the environmental impact of a column supporting a beam made of high-volume fly-ash concrete and obtained similar fly ash. The reduction of the use of cement results to be a crucial task for the organization of a useful strategy to save CO₂ emissions. Daminieli *et al.* [11] proposed a cement indicator named *binder intensity* (bi) index which defines the binder content required to obtain 1 MPa of mechanical strength, and consequently it expresses the efficiency of the binder material. The same authors also define a *CO₂ intensity index* (ci) which allows the estimation of the global warming potential of concrete composites. In concrete mix design, the clinker can be substituted by mineral additions, able to reduce its environmental impact. The tensile strength can be considered roughly proportional to the compressive strength [12] and, consequently, increasing values of this parameter produces greater emissions of carbon dioxide [10]. High tensile strength is also a primary engineering property to achieve durable and sustainable asphalt pavements [13]. In addition to compression and tensile strength, the ductility also plays a key role in the realization of high-performance concrete, because it provides significant improvements in the structural design life, further supporting the concept of environmental sustainability of such material [14, 15]. Such capability allows to limit the cracking propagation in concrete structural applications and sudden, explosive brittle failure. Recently, the development of micro- and nano-technology paved the way for new multifunctional high-performance materials with engineered particles, but in this field, the benefit/cost ratio in terms of energetic and environmental impact still needs further investigation [16]. Chiaia *et al.* [17] proposed a new *eco-mechanical index* (EMI) to quantify the environmental impact and the material performance of concrete structures. It takes into account strength, durability, fracture energy, water need, energy consumption

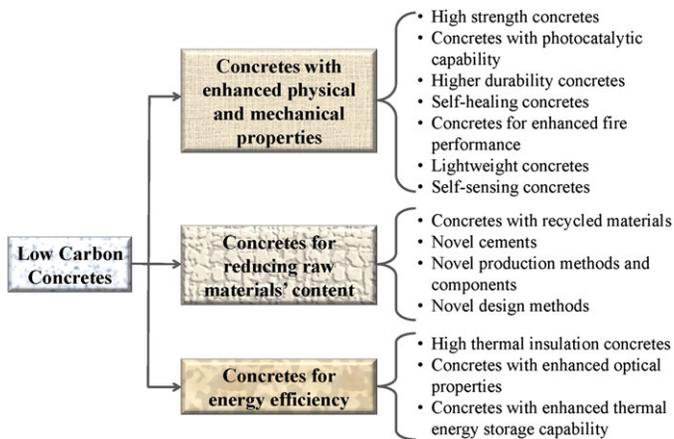


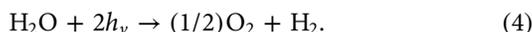
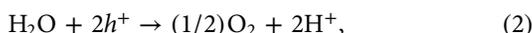
Figure 1. Conceptual scheme of the review.

and the amount of CO₂ emitted during the course of the concrete production process.

2.2 Concretes with photocatalytic capability

Semiconductors are materials which act as insulators at very low temperature, but possess a non-negligible electric conductivity at ambient temperature [18]. In practice, semiconductors are materials with an energy gap between the valence and the conduction band that is smaller than that of an insulator, such that, at room temperature, a significant number of electrons can be excited to cross the band itself [18].

When an electromagnetic radiation with energy equal or higher to that of the band gap ($h\nu \geq E_{bg}$) is absorbed by the semiconductor, an electron-hole pair is formed (case of TiO₂ in eq. 1). Such pairs can be used to produce electric energy (solar cells), to initiate a chemical reaction (photochemical catalysis) or even to produce an effective change on the material surface (superhydrophilicity) [19–21]. The former application, also known as photo-electrochemistry, was the first one to be investigated. It was developed in analogy with natural photosynthesis, and it is basically associated with the oxidation of water which determines the production of hydrogen (eq. 2–4 [22]). In this case, in order to avoid premature recombination of the photo-generated electron-hole pairs, an electric field is produced within the semiconductor, which drives the aforementioned pairs in opposite directions and enables to efficiently produce electric energy.



Superhydrophilicity, however, is the more recently discovered phenomenon associated with semiconductors in this field. Also

in this case, electrons and holes are produced, but the electrons tend to reduce the original substrate, while the holes tend to oxidize it, causing the ejection of oxygen atoms. The so-formed oxygen vacancies attract water molecules, triggering the semiconductor surface to produce OH groups (Figure 2), and become hydrophilic [23, 24].

Superhydrophilicity always occurs simultaneously with the last effect associated with semiconductors: photochemical catalysis. In this case, the photo-generated electron-hole pairs interact with molecular oxygen (O₂) and water (H₂O), producing two types of rather reactive radicals: superoxide radical anions (O₂⁻) and hydroxyl radicals (OH⁻), respectively. These two types of radicals were firstly found to be responsible for the decomposition of cyanide in water by Frank and Bard [25, 26] in 1977. After this revolutionary finding, a growing interest was registered in the possible environmental applications of semiconductors in air- and water-purifying photocatalytic reactions [27–30]. In particular, several studies focused on the removal of organic (VOCs, eq. 5 [20]) and inorganic pollutants generated within urban and industrial areas, which are ever more saturated by several substances emitted by anthropogenic sources.



These substances are responsible for many environmental issues such as air quality degradation, global warming and climate change. In this view, the use of photocatalytic air detoxification is a very attractive technique when applied to building surfaces exposed to these pollutants, since it spontaneously occurs at room temperature when even a small amount of UV-light with the right amount of energy interacts with a photocatalytic surface, without consuming auxiliary fuels, like in thermal incineration.

In principle, various types of sulfides and oxides can be used as catalysts for this kind of reactions (CdS, ZnS, TiO₂, ZnO, CeO₂, etc.) but the best photocatalytic performance is generally associated with titanium dioxide molecules (TiO₂) [20]. Titanium dioxide is the fourth most abundant metal on Earth,

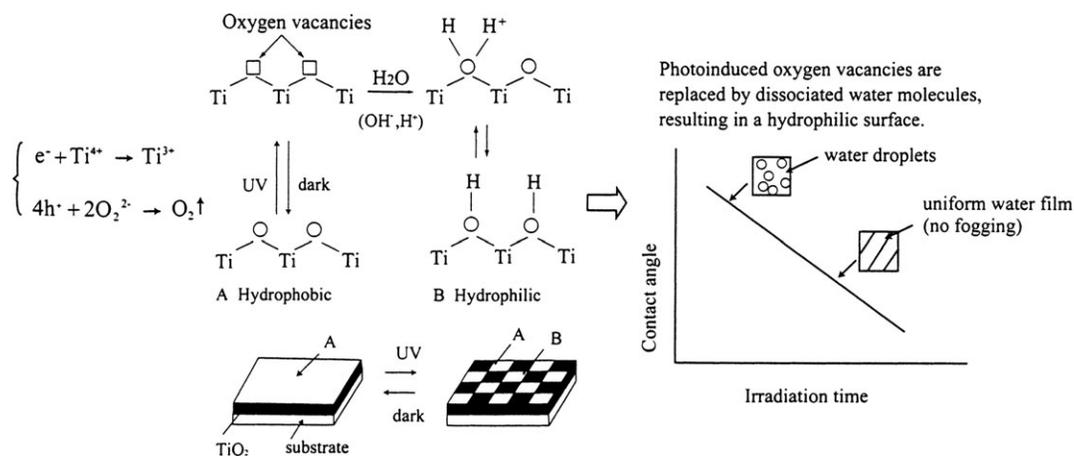


Figure 2. Mechanism of photo-induced hydrophilicity [25].

and can appear in different forms (anatase, rutile, brookite, etc.). Anatase and rutile, given their highest photocatalytic activity, are the most commonly used ones for this kind of applications [30]. Moving from the fundamental research to material science, titanium dioxide was effectively used in various fields. One of the most interesting ones definitely is associated with the production of cement-based building materials. In particular, the combination of TiO_2 with cementitious materials can have a 2-fold positive effect: the superhydrophilic phenomenon associated with titanium dioxide can produce smart building blocks with self-cleaning properties, while the development of superficial photocatalytic reactions can produce a significant reduction in both indoor and outdoor air pollution concentration (Figure 3) [31].

Titanium dioxide was effectively used in concrete products both as additive [34–36] and as a coating layer [37–39]. Most of these studies focused on the in-lab characterization of such material. In particular, Ballari *et al.* developed a kinetic model describing the degradation of NO_x molecules by concrete paving containing TiO_2 [40]. These models demonstrated to be in good agreement with the experimental results. Sikkema *et al.* and Ballari *et al.*, however, thoroughly investigated the effect of different parameters like NO_x concentration, UV-irradiance intensity and relative humidity, on the effective removal of NO_x molecules by TiO_2 -additivated concrete [41, 42]. Results showed a positive correlation with the nitrogen oxides concentration and the irradiance intensity, and anti-correlation with the relative humidity.

Interesting applications also evaluated the possibility to create specific TiO_2 -concrete mix designs using waste materials such as glass to enhance the photocatalytic effect. In this context, Poon *et al.* and Chen *et al.* demonstrated that the presence of recycled glass, with its high light transmittance properties, improved the quality of the photocatalytic effect [43, 44]. Furthermore, such studies also demonstrated that even though the glass size may not play a significant role in the catalytic activity, the glass color definitely does influence the intensity of the reaction. The effect of the different kinds of TiO_2 inclusion in the cementitious matrix was analyzed by Qin *et al.* [45], while Ramirez *et al.* examined the influence of two different

coating methods, i.e. dip coating and sol-gel process, on the toluene (VOCs) degradation capability of coated cementitious materials [46].

Despite all these interesting in-lab positive results, real-scale applications can highly change the boundary conditions and deeply influence the photocatalytic reaction. In this context, Chen *et al.* [47] compared the NO_x detoxification rate of in-lab and real-scale experimental campaigns. They found that the high variation of UV-intensity and temperature can significantly affect the reaction rate, causing a slight reduction on the NO_x decontamination potentiality with respect to similar lab-scale characterizations.

2.3 Higher durability concretes

The analysis of the literature reveals that much interest has been devoted to the compressive strength of concretes, while comparatively less investigated is their durability [48]. On the contrary, durability is an essential property to design concretes that keep their nominal strength during the lifetime of the construction under its environmental conditions. Indeed, when concrete elements are exposed to severe environmental boundary conditions, the durability of the structural material is of great importance [49] for structural safety and for the environmental sustainability of constructions which is affected by life-cycle maintenance and repair activities. Critical behavior results in concrete performance due to the presence of carbonation-induced reinforcement corrosion and freeze-thaw cycles. Proske *et al.* [49] carried out a study about concretes with reduced water and cement contents. They investigated compressive strength, workability and durability. The conventional cement of the reference concrete was progressively reduced from 270 to 100 kg/m^3 . Also fillers and additives were utilized gradually in substitution to the cement. A reduction of up to 35% of emissions was measured compared with reference concrete and of more than 60% with granulated blast-furnace slag.

The results of the research carried out by Limbachiya *et al.* [50] showed that binary and ternary cements have lower environmental impact with respect to equal design strength concrete. In particular, the finer cementitious components used in binary

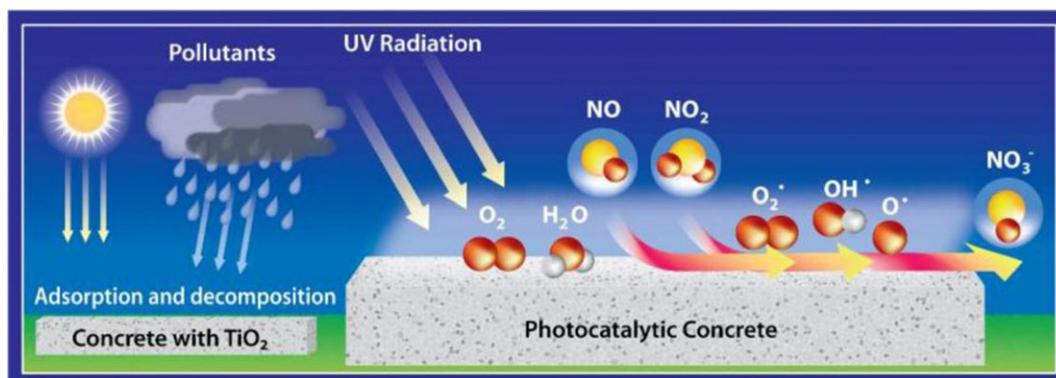


Figure 3. Schematic behavior of photocatalytic air purifying concrete pavement [32, 33].

and ternary systems in concrete production enhance the density resulting in enhanced durability performance. The research regarded both mechanical properties, such as compressive and flexural strength, and durability characteristics, such as initial surface absorption and carbonation. The experimental results of such eco-concretes, comparable to ordinary concretes, highlighted that they are suitable for structural use.

Hussain *et al.* review in [51] the state of knowledge on the durability of mortar and concrete containing alkali-activated binder with the aforementioned pozzolanic materials. They have the potentiality to attain good mechanical properties at early ages of curing with limited energy consumption and CO₂ emission. The durability properties of concrete and mortar with pozzolans and alkali make them potentially suitable for effluent, water and sewerage treatment plant. However, their characterization still needs deeper investigation before meeting the real market requirement.

Toledo Fihlo *et al.* [52] developed a sustainable Ultra High Performance Fiber Reinforced Cement Composite (UHPFRCC) produced using blast-furnace slag cement, silica fume, silica flour, wollastonite, steel fibers and superplasticizers. Compared with standard concretes and other UHPCs, they showed excellent performances in terms of rheology, mechanical performance and durability. Consequently, in UHPFRCC, an effective protection of rebars from chloride penetration and corrosion can be achieved with values of rebar cover thickness that are significantly smaller than those typically adopted when using standard concretes. The eco-friendly concretes investigated in the same research, mentioned above, seem to be suitable for structural applications, especially in the case of constructions that are particularly exposed to extreme conditions, such as roads, nuclear power plants, maritime structures, floors affected by abrasion and more. Reis *et al.* [53] proposed a sustainable concrete, realized with high volume of fly ash and they studied its experimental performance in particular with respect to its durability. Mixtures with high fly ash content exhibit low-embodied energy and greenhouse gas emissions [54]. Optimized concretes with fly ash exhibited excellent performances in terms of mechanical strength, resistance to chloride-ion penetration and resistance to freeze–thaw cycles. In general, the component characteristics and the mix design peculiarities of concrete could have more influence on the durability, in comparison to the traditional physical aspects [55]. Banthia *et al.* [56] presented the results of a research about the development of a sustainable concrete with fibers (fiber-reinforced concrete, FRC) particularly resistant to the deterioration. Results demonstrated that fiber reinforcements enhance the performance of normal concretes with respect to permeability under stress, bulk chloride diffusion, control of rebar corrosion and bond.

2.4 Self-healing concretes

Self-healing is the capacity of a material to bond the cracks when damages happen. Self-healing systems with microencapsulated healing agents primarily concern polymeric materials,

while the application of such technology to cementitious materials is relatively new [57]. Self-repairing is a mechanism of growing interest because it contributes to a longer service life of concrete structures making the material more durable and also more sustainable [58]. In general, self-healing concrete is classified into autogenous and autonomous healing [59]. Bacterial-based self-healing is a promising solution for sustainable concrete maintenance [60].

Wang *et al.* investigated the performance of encapsulated bacterial spores for self-healing concrete applications [61]. The water permeability of concretes with bio-microcapsules was ~10 times lower than that of normal concretes. Xu and Yao [62] investigated the self-healing properties of concretes with incorporated bacteria and calcium source nutrients, through mechanical tests at the macroscale (flexural and ultrasonic pulse velocity) and at the nanoscale (nanoindentation). The modified concretes showed a considerable impact on the healing efficiency.

Wang *et al.* [63] studied cementitious materials with hydrogel-encapsulated spores. Bacterial spores were encapsulated into hydrogels and then added to cement-based samples. The maximum observed healed crack width was ~0.5 mm and the water permeability showed a decrease of ~68%. Bacterial-based self-healing resulted promising for sustainable concrete maintenance.

To contribute to the development of a new generation of highly durable, damage-tolerant structures were proposed. As part of a class of cement-based composites exhibiting strain-hardening response, engineered cementitious composites have great potentialities for intrinsic self-healing due to tight crack widths and to the presence of additional cementitious materials in their mixes. In summary, they are promising for highly durable and damage-tolerant structures [64].

Sierra-Beltran *et al.* [65] investigated the durability problems of currently available concrete repair systems and proposed the use of strain-hardening cement-based composites with addition of bio-based agents. They evaluated their mechanical properties and the bonding behavior with the concrete substrate.

2.5 Concrete for enhanced fire performance

Section 2.1 introduced the concept of HSC, whereby the enhanced mechanical performance of the material allows to address low-carbon target by reducing the size of structural members to withstand similar loads. HSCs are widely used in massive and innovative structural designs such as off-shore structures, large bridges and infrastructural systems but, lately, their application has spread toward building columns even in standard constructions. These structural members can be considered as the main load-bearing components of the building envelope, and for this reason, their fire resistance deserves a special attention. Although HSC columns can claim enhanced mechanical properties in common conditions, a vast body of literature, starting from the initial contribution by Hertz in 1984 [66], demonstrated a huge decrease of such mechanical properties under fire conditions [67–69]. When the structural member

is exposed to a rapid temperature increase, in fact, normal strength concrete (NSC) presents better performance when compared to HSC. This huge decrease in the mechanical resistance in HSC structural elements is basically due to the occurrence of the spalling effect. Spalling can be considered as the result of two different but concomitant processes: thermomechanical and thermohydral process [70]. The first process concerns the occurring of thermal dilatation/shrinkage gradients within the heated structural member [71, 72], while the second one is associated with the generation of high-pressure fields of water vapor and enclosed air, in the porous cementitious matrix [66–73]. Both phenomena can be associated with the high reduction of permeability characterizing HSCs. In this context, the scientific literature enumerates a large number of studies trying to identify the fundamental factors affecting the mechanical resistance of HSCs and to minimize the spalling in such structures in order to obtain enhanced mechanical resistance also in case of fire conditions. Therefore, it is possible to state that the fire performance of a concrete column can be influenced by different factors like moisture content, concrete density, fire intensity, presence of silica fumes, lateral confinement, member dimension, load intensity, loading type and more [73]. In general, higher moisture content, which is also a consequence of lower density in HSC, is associated with higher vapor pressure and spalling. Furthermore, a fast heating fire, together with a massive dimension of the structural element and the presence of silica fumes, enhances the risk of explosive spalling. Nevertheless, lateral confinement and the presence of low amount of additional loads seem to ensure a better fire resistance and the material durability in case of fire [69, 73–75].

Starting from all of these considerations, different kinds of additivated HSCs were developed in recent years. The introduction of polypropylene (PP) fibers within the cementitious matrix can be considered as one of the most successful implementations of HSC. The reason for this high contribution to fire resistance is probably associated with the very low melting temperature of such fibers, i.e. 170°C. Once these fibers are melt, in fact, free channels are created for steam pressure within the HSC column, thus preventing local high vapor pressures, which generally cause spalling.

Various research contributions reveal a significant reduction of the spalling effect in HSC columns because of PP fibers addition [73, 76–79]. A lot of these studies also investigate the optimum concentration of PP fibers and their most suitable length for concrete applications. Results show that the enhanced HSC behavior can be reached with a very low percentage of fibers, i.e. 0.1–0.15% by volume. Furthermore, the introduction of Nylon (NY) fibers, which are characterized by smaller dimensions, can reduce this fiber percentage up to 0.05%, i.e. 0.025% of PP and 0.025% of NY fibers, respectively [80]. This further reduction is very important, since it is associated with a better workability of the mixture, and consistent spalling reduction effect.

Another kind of material that seems to produce a beneficial effect on the HSC is constituted by steel fibers, which are able to increase the tensile strength of the compound and reduce the

spalling effect [73]. As for the main fiber dimension, different research works demonstrate that it should be selected with respect to the inter-aggregate spacing of coarse aggregates in the concrete mixture [81, 82]. The fire resistance of HSC columns also seems to be positively influenced by the use of carbonate aggregates in the mix design (generally limestone) which, given their higher specific heat values, can produce a further 10% increase in the time leading to failure during a fire event, when compared to siliceous aggregates (mostly quartz) [77, 83]. Lastly, when the spalling effect cannot be avoided, the use of properly designed transverse reinforcement for lateral confinement can at least ensure a reduction of its effect. In particular, an improved configuration of stirrups, with adequate connections to the internal core of the column, and a reduced stirrups spacing can be highly beneficial [73].

2.6 Lightweight concretes

LWC is a type of concrete used for low-density structural and non-structural elements [84]. The influential factors for improving the sustainability of LWCs is due to their relatively low weight, environmentally friendly behavior and their easy industrializing and on-site casting [85]. A more sustainable LWC can be obtained by substituting the traditional raw aggregates with recycled wastes coming from other industrial activities. The results of this research showed reduced permeability of the concrete due to the use of fly ash or silica fume, and improved mechanical properties due to palm oil fuel ash as a pozzolan. Gunning *et al.* [85] propose a treatment of accelerated carbonation, in order to convert gaseous CO₂ into solid calcium carbonate through a reaction with industrial thermal residues. The obtained carbonated aggregates showed analogous properties compared to the traditional LWA's.

The use of lightweight and energy-saving cementitious materials is one of the interesting trends of modern architecture and building technology [86]. Indeed, LWC can reduce the structural dead loads, the loads of foundations, and consequently the construction costs. Lightweight aggregates can be also inorganic and low-embodied-carbon composites. Yang and Zhang [87] studied a new type of lightweight inorganic aggregate concrete by replacing organic material. That material is also energy efficient and energy saving.

Park *et al.* [88] investigated low-density/low-cost cement composites with carbon and alkali-resistant glass, i.e. fly ash with silica fume. Carbon fibers behave better than glass fibers, due to their superior chemical and mechanical properties. Thermosetting plastics, which cannot be melted by heating and reused as new plastic, can be utilized for non-structural LWC [89]. The addition of structural fibers improved compressive strength and post-peak ductility of cellular LWC masonry [90]. Micro-fibers (fibrillated) enhance pre-cracking behavior of masonry by arresting cracks at micro-scale, while macro (structural) fibers induce ductile behavior in the post-peak region by arresting the crack propagation relatively sooner after the crack initiation.

2.7 Self-sensing concretes

Self-sensing concrete is able to identify variations of applied loads or displacements, temperature or damages without the use of external traditional transducers [91]. The self-monitoring ability is obtained through the dispersion of engineered electrically conductive fillers into the cementitious matrix [92]. The changes of external strains and stresses are converted into changes of electrical properties, such as electrical resistivity or conductivity. The fillers can be made of carbon-based, metallic or polymer materials [93]. Some conductive fillers, including hybrid combinations of different types of fillers, are also able to enhance the major mechanical properties of concrete, such as compressive strength and durability [94, 95]. For the achievement of an optimal conductive network, the choice of the filler concentration and the dispersion method are key parameters affecting the property of self-sensing concrete [96]. Self-sensing concrete is suitable for the construction of structures and infrastructures with multifunctional properties [97, 98]. Self-sensing concretes can allow an automated identification of anomalies in the structural behavior, for instance associated with a developing damage pattern, thus providing valuable information for cost-effective maintenance and restoration activities [99–102].

Future promising applications can be represented by the monitoring of concrete and asphalt floors, traffic and crowd control, active and semi-active structural control, thermal and hygroscopic sensors for environmental management of structures and high-energy efficiency concrete, thanks to the addition of conductive nanofillers and phase change materials [103–105]. Self-sensing concrete can be considered as a promising sustainable instrument, for its high performances, for the enhancement of the durability and the safety of the structures, for the decrease of the city's emissions coming from a correct management of the traffic [106].

3 CONCRETES FOR REDUCING RAW MATERIALS' CONTENT

3.1 Concretes with recycled materials

The construction sector is notoriously associated with a huge environmental impact mostly brought upon by the production of growing quantities of concrete. Cement-manufacturing, in fact, is an extremely energy-extensive process, which results in the emission of a huge amount of greenhouse gases, while the excessive usage of aggregates is associated with a massive depletion of natural resources. In addition, all human activities apart from concrete production produce a huge amount of waste materials and by-products, whose management has nowadays become a major environmental concern. Extensive consumption of natural resources and huge production of industrial wastes need to be dealt with in order to reach what is called sustainable development.

In recent years, a promising strategy consisting in the reutilization of waste materials and by-products in construction

building materials has been proposed. As a result of this approach, waste materials and by-products are effectively reintroduced in the production chain, and the depletion of raw materials for producing concrete and other building materials is highly reduced. Furthermore, some of these materials, when properly treated and used, may even produce an improvement on the mechanical and durability properties of mortars and concretes, which could not be achieved by simply using ordinary Portland cement. Given the promising advantages of such combination, research studies on cement and concrete materials all over the world are currently exploring an enormous number of combinations to develop cost- and resource-effective recycled concretes.

In this review article, recycled concretes are classified as by-products concretes and waste materials concrete, where a by-product is identified as a secondary product derived from a manufacturing process or a chemical reaction, while a waste material is considered as an unwanted or unusable material, which is discarded after primary use [107].

3.1.1 By-products concretes

In theory, all the production processes or chemical reactions are associated with the creation of by-products characterized by very different properties and compositions. The concept of using industrial by-products for the production of concrete is a well-established one. It dates back to the 1970s, when silica fumes were firstly used as additives to create dense concretes with enhanced mechanical properties [108]. Silica fume, also called super-pozzolan, consists in 90% of SiO₂ and it is widely used as a replacement for Portland cement, i.e. up to 9–15% by cement mass content [109]. The use of silica fume was thoroughly investigated in a huge number of research works [110, 111] and its principal advantages consist of the increased pore refinement and the improved strength.

Two additional fine-matter industrial by-products, namely fly ash and slag are also successfully used as supplementary cementitious material (SCM) in the production of mortars and concretes. Fly ash is one of the coal combustion products, while slag is a secondary product of the iron manufacturing process in a blast furnace. Both materials are characterized by a silica-alumina content which can be considered as similar to that of pozzolanic materials and exhibit an interesting pozzolanic activity [112]. Their use as cement substitutes modifies the cementitious matrix and the pore structure, producing hardened cementitious composites [113–116].

Lastly, industrial processes are also very often associated with the generation of coarser by-products which can be used as aggregates in the production of LWCs with enhanced insulation properties. Among these products, wood shavings and paperboard mill residues [117] showed interesting mechanical properties and durability, and can be considered as suitable elements for the production of LWC [118, 119].

3.1.2 Waste materials concretes

Three different classes of waste materials can be identified for the production of recycled concretes: (i) demolition wastes, (ii)

industrial wastes and (iii) agricultural wastes. Each one of them is dealt with in the following subsections of this work.

Demolition waste products As it is well known, concrete is the most widespread construction material worldwide, but it is not an everlasting one, which stimulates life-cycle considerations. The industrialization encourages the development of construction and demolition activities and consequently the generation of increasing amounts of debris and waste [120]. In this context, it is clear that being able to effectively reuse wastes from a structure demolition could represent a way toward a more sustainable use of such material. Because of the promising applications of such demolition wastes, their use as concrete aggregates are widely investigated by the scientific community, as discussed below.

Before being used to produce new concrete composites, demolition wastes are turned to small pieces, i.e. recycled concrete aggregates (RCA). Such aggregates do not exhibit the same characteristics of the original ones, since they generally cannot be separated from the mortar component that remains well attached to their surfaces, and present impurities such as glass, metal, gypsum or wood coming from the demolition process. Given this considerations, RCAs have been found to be of poor quality, when compared to the original product, being associated with higher porosity and lower density [121–123]. In addition, the weak quality of the bond between the original aggregate and the attached mortar residue, together with the presence of small cracks caused by the crushing process and the highly dispersed particle size of the RCA seem to produce a further enhancement in porosity and decrease in mechanical strength in the final recycled concrete [124, 125].

In light of the considerations made above, RCA-concretes realized with even a partial substitution of primary aggregates with the recycled ones produce a general decrease in their mechanical properties [126]. Nevertheless, different research studies proved that the use of specific pre-treatments procedures can highly reduce such harmful decrease, i.e. RCA coating procedure, impurities removal procedure, oven curing technique and RCA calcination [124, 127–129].

Industrial waste products The use of industrial wastes to produce eco-concretes is receiving a considerable interest and can rely on a massive amount of possible composite candidates with completely different characteristics. The research panorama is thus fully populated by heterogeneous applications which investigate a variety of composites. This review article will only consider the most acknowledged ones like those associated with the reuse of plastic materials and tire rubber.

Both the aforementioned wastes are characterized by a low biodegradability and the need of large land areas for their storage. Furthermore, the method of recycling such materials in concrete composites is not the only possible one, but it can effectively contribute to an efficient final disposal.

The use of plastic waste materials in concrete has been tackled by considering a wide range of possible waste forms,

e.g. plastic coarse aggregates [130], waste plastic flakes [131], granulated plastic waste [132], polyvinyl chloride [133], etc. Despite the huge number of concretes associated with plastic polymorphous nature, the current research background seems to denote similar properties in a large variety of the investigated composites. Firstly, both tensile and flexural strength of the different mixtures improve because of the bridging and the crack arrester effect, respectively [134, 135]. Also, compressive strength decreases with increased plastic content [136]. Furthermore, the modulus of elasticity of concrete and its workability generally decrease with increasing plastic content in the mixture [137].

The second kind of industrial waste material presented in this section is the tire rubber. The use of such material in combination with Portland cement concrete is motivated by the notable results obtained in the production of crumb rubber asphalt paving. The coupling of these materials, in fact, gave birth to an asphalt mixture with improved performance, given their intrinsic mutual compatibility.

Despite the acknowledged success in the combination of rubber and asphalt, results from different research studies seem to show that the combination of rubber wastes and concrete matrix is not as effective as the previous one. Generally, a significant reduction of compressive strength and split tensile strength can be noticed, together with a significant enhancement of the mixture toughness and fracture resistance [138–140]. The reasons for the poor behavior of the investigated composites are described as a combination of two different factors: the presence of stress concentration due to the significant variation in stiffness, and the intrinsic chemical incompatibility of the considered materials [141–143]. This field of research, however, is still working on the implementation of effective solutions to reduce the strength loss while keeping the beneficial effects of the rubber-based concrete.

Beside plastic and rubber, further waste materials were investigated in concrete mixtures with the main purpose to produce eco-concretes, possibly with enhanced mechanical properties as well. Among these materials, ceramic and glass ones are important enough to be noticed. The former class of materials, when combined with concretes, seems to produce no adverse effect on its consistency, while it increases its mechanical properties [144]. The latter, however, is giving interesting results in autoclaved aerated concretes, where it produces composite materials with similar mechanical properties compared to the original ones [145].

Agricultural waste products The last class of considered concretes with waste materials is the one associated with agricultural residues. Given their widespread distribution all around the world, this kind of eco-composites is currently being studied with a deep interest by the scientific community. Just like in the case of industrial wastes, the massive variety of such agricultural residues determines the possibility of creating a large amount of composites with interesting thermomechanical properties. In general, agricultural wastes can be combined with

concrete in three different ways: they can be used as partial cement replacement material, partial aggregate replacement or fiber reinforcement for concrete composites. The former application is justified by the high amount of minerals and silicates that an annually grown plant can absorb from the earth during its growth process. Such a chemical composition, in fact, can yield to a material with strong pozzolanic reactivity, which is a representative characteristic of cement replacement materials [146]. The use of natural FRC, however, can basically be considered as an alternative to the use of synthetic ones. Such an alternative seems to be very promising since natural fibers combine high mechanical properties, i.e. similar to those of the most common artificial products, with low costs and environmental impacts [147]. Lastly, using agricultural wastes as a partial replacement of aggregates in the concrete mixture seems to be an encouraging way to reduce the dependence on common materials such as gravels and natural mining sand [148].

The most common agricultural wastes used to produce cement replacements are the ones also used for other building materials, such as sugarcane bagasse (which is actually a by-product of the sugar extraction process but it is here described with the other agricultural wastes), common reed, rice husk ash, wheat straw ash, bamboo leaf ash and corn cob ash [150–153]. All these materials are turned to ash by means of a combustion process: they are generally burnt in a furnace or an incinerator in a controlled atmosphere with precise temperature ranges which can vary from 600 to 1000°C, depending on the specific waste. When such ashes are partially substituted to cement in the concrete preparation, they generally produce a porosity decrease and an enhancement of mechanical strength. However, the increase on the mixture water absorption causes workability issues, which highly restrict the maximum content of waste material to be used in the final mixture [151–156]. The substitution of common aggregates with agricultural wastes, mostly represented by wheat, corn and olive crops, is a very promising application. Also in this case, the mixture increases its water demand and reduces its workability, but the results in terms of mechanical properties are very interesting especially for wheat particulate, which when combined with limestone produces an impressive increase in compression, tensile and flexural strength of autoclaved concretes, i.e. 87%, 67% and 71%, respectively [157, 158].

The last kind of agricultural waste concrete which has been recently investigated in a huge number of research works is represented by vegetable fibers. The commonly used fibers are hemp, coconut shell, bamboo, wheat, corn and sisal fibers. Hemp-concrete is a well-established LWC with impressive insulation properties (thus described in more details in Section 4.5). It is generally produced using lime (possibly natural hydraulic lime, sand, pozzolans and/or cement) and its density corresponds to about one-seventh of that of ordinary concrete. It lacks of brittleness and it is associated with a typical compressive strength value of ~1 MPa, which does not allow to use hemp concrete as a structural material [159, 160]. All the other fiber concretes can be used to produce LWCs with enhanced

flexural strength, but generally reduced compressive strength. Nevertheless, the mechanical characteristics of such composites are highly influenced by two main parameters: the main tensile strength of the fiber itself, and the bonding efficiency between the fiber and the cementitious matrix. Failure in FRC can either occur for rupture or pull-out of the fiber, depending of the aforementioned parameters [161–163].

3.2 Novel cements

Environmental impact of cement production can be at a global or a local scale [164]. At the global scale, the CO₂ emissions can be reduced by alternative clinker chemistries. Low-energy belite cements could be a valuable alternative. Local environmental impacts include SO₂ and NO_x emissions which contribute to acid rain and cement kiln dust emissions. Environmental and health risks can be significantly reduced by means of alternative fuels and mineral carbonation [6].

Randl *et al.* [9] emphasized the importance of new mixes for UHPC, substituting cement with high environmental impact with local available alternative materials so as to achieve a ‘green’ concrete. The developments of cement and concrete green technology involves new materials and production methods [165]. The production of novel cements concerns the modification of several processes that use various raw materials. The ideal novel cement emits less CO₂ and requires less energy to produce Portland cement, without significantly affecting its efficiency. Ideal cements would preferably use waste from fuels and raw materials. Existing candidates of resource efficient cements are available and new ones are developing [166]: calcium sulfoaluminate cement [167], calcium aluminate [168], super-sulfates cements [169], magnesium oxide-based cements [170], alkali-activated cements [171], geopolymers [172] and sequestered carbon cement [173].

Gartner and Hirao [174] proposed, in the range of alternative approaches for reducing greenhouse gases emissions, the replacement of Portland clinker in concrete with low-carbon SCM, inserted with the cement or directly into the concrete mixture.

Binary and ternary cements, that are conventional Portland cements with addition of one or two SCM, such as fly ash, ground granulated blast-furnace slag and silica fume, have been recently investigated in [4], showing that such special cements result in a reduction of the environmental impact of concrete constructions for a given design strength and regardless of the increased cementitious content.

The use of novel cements needs a standardization [175]: many countries developed national concepts and defined the possible cement components and types for the realization of concrete for constructions.

3.3 Novel production methods and components

In energy analysis, the embodied energy is related also to the subsidiary processes with respect to the final product [172]. Building materials with high-embodied energy in general result

in more environmental impact than would materials with low-embodied energy [176]. The development of new energy-efficiency technologies in research works and in the market represents a mid- and long-term climate protection strategy in the field of cement and concrete [177]. Several emerging technologies are available or in a phase of study, aimed at reducing the energy consumption and the carbon dioxide emissions in the various steps of the cement production [178]. The main emerging grinding technology is the high activation grinding that increases the presence of additives in the cement and concrete production [179]. Hasanbeigi *et al.* [177] described also other 17 emerging technologies in energy saving: fluidized bed kiln [180], calcareous oil shale as an alternative raw material [181], the use of steel slag as kiln raw material [182], non-carbonated raw material for cement production [183], cement with low lime saturation factor [184], cement and construction materials based on magnesium oxide [185], geopolymers cement [186], cement primarily of fly ash and recycled materials [187], capturing the CO₂ resulting from limestone precalcination [188], CO₂ sequestration in concrete curing technology [189], carbonate looping technology [190], bio-technological carbon capture [191], oxy-fuel technology [192], post-combustion carbon capture using absorption technologies [193], Calera process [194], industrial recycling of CO₂ from cement process into high-energy algal biomass [195] and, finally, the use of nano-technology in cement and concrete production [196].

Gartner and Hirao [168] identified as possible modalities of reducing the CO₂ emissions in the cement industry the use of alternative fuels (including tires, sludge, waste oil, plastic and biomasses) and the capture and sequestration of the gas by cement plants. Some capture technologies appear more suitable for the application to the cement industry [197]: oxy-fuel technology, post-combustion capture, mineralization of CO₂ in an aqueous precipitation process and the use of photosynthesis of algae.

Thermal and electrical efficiency of cement plant can be enhanced with retrofitting devices aiming at avoiding energy loss and great consumption of fossil fuels; unfortunately, this solution is not frequently used because of its cost [198].

Great potential for reducing the CO₂ emissions is for NSC. There are several possible approaches regarding the modification of the mix design: the introduction of superplasticizers and of more reactive cements, the decrease of the water content, the optimization of aggregate-size distribution and the use of mineral inclusions [199]. In this way, the Portland clinker can be reduced. In addition, the introduction of fly ash or furnace-slag in the clinker, coming from the recycling, has the double utility in the limitation of the use of natural resources.

Carbon dioxide emissions related to concretes are mainly associated with the production of Portland cement. The partial substitution of the cement with less-impacting materials, coming from other industrial processes, is a practice of growing interest for the sustainable protection [200]. At the same time, the substitution of virgin aggregates with different recycled materials is increasing with a consequent gain for the environment. The most important additives for concretes related to

these two types of substitution are fly ash, ground granulated blast-furnace slag, silica fume, recycled concrete, post-consumer glass, recycled tires and plastics, and by-products of the article and other industries.

3.4 Novel design methods

In order to minimize carbon emissions, it is essential to produce building materials and products with minimum amount of energy need for their installation and transportation. Some alternative constructive solutions can be realized with less energy consumption compared to others [201].

In traditional structural design, just a weak attention is paid to environmental issues, which gain significance in modern integrated design approach. The Semantic Web technologies represent an innovative tool for structural engineers in enhancing constructions' sustainability [202]. The selected sustainability indicators are the embodied energy and CO₂. In other proposed design methodologies [203], the experimental design variables were the type of binder and concrete, the area of steel, the diffusivity, the concrete cover depth, which had an influence on the service life sustainability of concrete. The aim of this experimentation was to identify the optimal dimensions and material properties for an RC (reinforced concrete) beam with low environmental impact. Other studies regarded the cost and environmental impact optimization of flexibly formed concrete beams and of steel RC columns in high-rise buildings [204]. Therefore, the interest in environmental-friendly solutions in construction design is a topic of growing interest. In 2010, Masdar City announced a prized-competition for the best proposal of 'Sustainable Concrete' and 'Lowest Carbon Footprint' for buildings in the city with a total of 2 million cubic meter of concrete [206]. The results of an experimental campaign carried out by Elchalakani *et al.* showed that the slag concrete mixes considerably diminish the carbon footprint and meet the design requirements.

García-Segura *et al.* [207] proposed a method to design reinforced concrete I-beams based on multiobjective optimization techniques. The aims were to limit the design costs, the CO₂ emissions, without reducing the service life and the safety coefficient. The methodology enhances the sustainability of the design and is applicable to various structures.

Yeo and Potra [208] developed a novel optimization approach of decision-making in order to achieve sustainability and economic results. New optimization instruments are indeed available to perform efficiently large volumes of calculations applicable to several structural engineering problems. For structures subjected mainly to large compressive forces, such as high-rise buildings, the reduction of the CO₂ footprint achieved by optimizing the design to achieve minimum carbon emissions can be considerable. As highlighted by multiple recent effort in this field, modern and multifunctional constructions need to be definitively environmentally sustainable, healthy, smart and technologically advanced by taking into account the whole life-cycle impact of their development [209].

4 CONCRETES FOR ENERGY EFFICIENCY

4.1 High-thermal insulation concretes

From a merely energetic point of view, LWC, thoroughly described in Section 2.6, can be considered as a particular kind of concrete with an enhanced thermal insulation capability. Such an enhancement is mainly due to the introduction of a gas or a foam agent within the concrete matrix, or to a partial replacement of ordinary aggregates with low weight, and possibly cost-effective ones [210–212].

The thermal conductivity of an ordinary concrete generally varies between 0.62 and 3.3 W/mK, depending on the type of aggregate and the hygrothermal conditions of the sample [213–215]. When a common LWC is considered, however, the lambda value generally varies in the range 0.4–1.89 W/mK [216, 217] and reaches even lower values when polystyrene beads or foams are used within the mixture [218, 219].

In this review article, LWCs are divided in two different classes of materials: inorganic-based LWC and bio-based LWC. The first class of mixtures is composed by all those LWCs which owe their insulation characteristics to natural minerals and artificial materials, sometimes even recycled ones, while the second class is mainly composed by biological natural fibers or particulate.

4.1.1 Inorganic-based LWC

Using inorganic materials to produce LWC is a well-recognized procedure. Different research studies investigated the use of volcanic materials such as diatomite, pumice and basaltic lapilli [220–227], and observed their effect on both mechanical and thermal properties of the obtained LWC. In general, results showed that the high intrinsic porosity of such materials causes an appreciable thermal conductivity decrease in the final composite, which generally is found to possess higher insulation performance for higher percentage of volcanic aggregate. Unala *et al.* [220], for example, found that the use of different grain size of diatomite in combination with ordinary Portland cement can produce a minimum λ value of ~ 0.23 W/mK, while Sanchez Fajardo *et al.* [221] found that the thermal conductivity of LWCs produced using basaltic lapilli as aggregates drastically increases when the mixture is taken from a laboratory dry state to a water saturated condition.

The analysis of the literature also seems to reveal that thermal insulation properties similar to those obtained with natural volcanic stones can also be reached with using expanded clay, expanded shale and perlite [222, 223], while in order to achieve a significant decrease of this parameter, the use of EPS (expanded polystyrene) beads is required [224–226]. Furthermore, when such material is partially replaced with EPS foam, the thermal conductivity of the obtained concrete is found to reach the surprising value of ~ 0.06 W/mK while also enhancing the related mechanical properties [219].

It is noteworthy to mention a further kind of artificial inorganic aggregates used in the production of high insulation

concretes: industrial wastes such as plastic, glass, rubber and even EPS itself.

The integration of extremely low-density and low thermal conductivity ethyl vinyl acetate wastes in a common LWC, for example, enabled Duslang *et al.* [227] to slightly reduce the overall thermal conductivity of the investigated cementitious mixtures from 0.489 to 0.407 W/mK. Glass beads, however, were effectively used to reduce the thermal conductivity of insulating concrete by Chung *et al.* [228], while also analyzing the effect of voids interconnection on the overall thermal behavior of the sample. Lastly, the inclusion of rubber crumb in sand concrete blocks was shown to produce a global λ decrease of ~ 5 –10% on the final composite [229].

4.1.2 Bio-based LWC

The use of bio-based materials for the production of LWC is a rapidly growing field of research which enables to somehow reduce the depletion of raw materials and the harmful greenhouse gas emissions in the atmosphere, while addressing thermal energy efficiency in buildings. This 2-fold effect is mainly due to the inner nature of the bio-based material used to produce such concretes. Bio-based LWCs, in fact, use lignocellulosic material from agricultural or industrial wastes, which generally provides concrete with a high amount of interconnected porosity, resulting in a huge insulation capability. Two are the most common natural materials used in the production of such environmentally friendly mixtures: wood shavings and hemp.

Thermophysical properties of lightweight wood chipping concrete were assessed by a huge amount of studies [230–237]. Among such research works, Belhadj *et al.* [234] investigated the effect of the incorporation of barley straws and wood shavings on the physical–mechanical properties of sand concrete intended for constructions in arid zones, showing that the use of selected local materials and agricultural wastes can improve the thermal performance, while minimizing costs and energy consumption. Al Rim *et al.* [236] investigated the effect of different wood proportions on the thermal and mechanical performance of cement-wood composites finding thermal conductivity values ranging from 0.24 to 0.08, i.e. 10% and 50% in weight of wood, respectively, while Taoukil *et al.* [237] examined the effect of moisture content on the thermal conductivity and diffusivity of wood-concrete composites, finding huge discrepancies with increasing relative humidity [238]. The same authors also proposed to somehow treat the wood matter in order to reduce the water absorption capacity, and with it the perturbation of thermal properties of the composite.

Hemp fiber concrete is the reference material when it comes to new bio-based LWC characterization [238–243]. In particular, lime/hemp lightweight blocks can be considered as rather conventional building materials with an overall thermal conductivity in the range of 0.1 W/mK [239]. Nevertheless, hemp has been proven to absorb a large amount of water, which generally causes long setting and drying times. Elfordy *et al.* [241] defined a new method to produce ‘hempcrete’ mostly consisting

in adding, just before the hose outlet, the amount of water strictly necessary to the slaking process of the lime. This particular method seems to drastically reduce the drying time given the accelerated kinetic setting, and produce a slight increase in thermal conductivity, modulus of elasticity, compressive and bending strength and hardness.

Beside the two most common applications of wood and hemp LWC, a significant attention has also been devoted to the study of thermal insulating properties of a huge amount of natural fibers less commonly associated with the building sector. Among such fibers the most interesting results are those related to cork, corn cob, sugarcane, cellulose loose-fil, flax, straw bales and coconut [Table 1].

4.2 Concretes with enhanced optical properties

Energy-saving concretes are also those using enhanced optical properties to reduce the overall building energy demand for cooling. Such applications focus on the use of 'cool' materials characterized by high solar reflectance, i.e. higher than 65%, and also high-thermal emittance values, i.e. around 0.80–0.90. Building envelopes components having such optical properties are able to reduce the amount of solar energy absorbed by the material, causing a drastic surface temperature decrease, and consequently an appreciable reduction in the bordering air temperature [244]. Additionally, if applied as urban paving materials, they can contribute to Urban Heat Island mitigation, by also improving the livability of the outdoor urban environments for pedestrians [245, 246]. In fact, since almost 50% of the solar energy reaching the hearth surface can be found in the infrared region of the solar spectrum, cool materials can be both constituted by light-colored white coatings, or highly reflective colored coatings [133–136, 243–249]. In particular, the amount of solar radiation actually reflected by light-colored coatings is either due to white pigments with solar reflectance values of ~0.70–0.85, i.e. zinc dioxide, titanium dioxide (generally in the form of rutile and anatase) [137, 138, 250, 251,] or transparent polymers such as acrylic. Colored-cool materials, however, exhibit the same solar reflectance of non-cool materials in the visible wavelength range, but ensure as high solar reflectance as the light-colored coatings in the near-infrared wavelength range.

Table 1. Density and thermal conductivity values of some natural fibers used to produce LWC [244].

Fiber raw material	Bulk density (kg/m ³)	Thermal conductivity λ (W/mK)
Cork	120–180	0.045
Corn cob panels		0.139
Sugarcane	100–125	0.0469–0.0496
Cellulose loose-fill		0.05
Flax	5–50	0.038–0.075
Cellulose (recycled paper)	30	0.041
Hemp	20–45	0.040–0.060
Straw bales	102.6	0.067
Coconut	85	0.058

Two further kinds of cool materials have been used to produce energy-efficient coatings for building applications: retro-reflective materials and thermochromic materials [252–260]. Retro-reflective materials are constituted by highly reflective coatings, including glass beaded type and cube corner type, able to reflect the incident solar radiation in the same direction of the incoming one. Such materials are particularly effective in decreasing the mutual radiative effect of close standing buildings, and in this way contribute to building energy efficiency [253, 254]. Lastly, thermo-chromic-doped coatings [255, 259] are an innovative kind of materials which can dynamically change their thermal–optical properties in response to varying environmental conditions. Such materials, in fact, using a reversible reaction of the pigments molecular structure, reflect most of the incoming radiation in summer conditions, thus reducing the associated building thermal load, and absorb most of this radiation in winter, when the internal heating needs significantly increase [257, 258].

Light-colored and colored-cool materials, retro-reflective and thermochromic materials, are generally used to produce films or coatings. Such coatings have been effectively applied to different kinds of concrete elements [257, 259], and their use as concrete coatings is reported to produce a significant reduction in the indoor operative temperature in warm/hot conditions [262].

Two are the most successful cool-coated concrete building elements: tiles and pavement blocks. The use of cool concrete tiles as roofing components of an innovative building has been widely acknowledged by the scientific community, which found the enhancement of the solar reflectance index associated with such composite materials to be very effective in reducing building energy consumption [258, 261, 262]. Pavement blocks, however, representing almost the 30% of the whole urban areas [263], have been highly appreciated for their contribution to the Urban Heat Island mitigation and the improvement of the outdoor environmental conditions [259, 264]. Of course, such environmental benefits produce a direct rebound effect on the amount of energy consumption associated with the building stock, thus leading to an overall reduction of building cooling needs. Generally, the implementation of high reflectance cool paints, either white or colored, has been shown to effectively increase the albedo of concrete blocks, which normally ranges between 0.4 and 0.2, for new and old concrete blocks, respectively [265–267]. Synnefa *et al.* [265], for example, applied such paintings on the surface of white concrete paving tiles and reached final albedo values between 0.8 and 0.9 and an emissivity higher than 0.8, for non-aluminum pigments coatings. Such enhanced optical properties caused a reduction of 4 and 2 K in the daily and the nightly surface temperature, respectively, in summer conditions in Athens [265]. A variety of further applications also showed the effectiveness of colored coatings in reducing the surface temperature of concrete paving [259, 260, 268]. In this context, Synnefa *et al.* [260] assessed a maximum daily peak temperature reduction in August of 10.2 K for the 'cool' black paving compared to the same standard black

finishing. Furthermore, the use of thermochromic paintings as coating layers for concrete pavements has been shown to be even more effective than high reflectance cool materials, but their proven rapid loss of optical characteristics represents a major problem for real-scale applications [269, 270]. Lastly, it is worthwhile to mention that special concrete mixtures, obtained by partially replacing cement with fly ash or slag, allowed to produce concrete pavements that were able to guarantee albedo values of ~ 0.6 , with a 70% replacement of cement with fly ash [271]. Such results are quite interesting, although the acknowledged performance and the simplicity of application of coated products strongly penalize the developments in this research field of high albedo concrete surfaces [246].

4.3 Concretes with enhanced thermal energy storage capability

Thermal energy storage systems allow to collect and store heat or cold and use them at some future date under varying temperature conditions. This particular energy-efficiency application focuses on the assessment of two different key points, i.e. overcoming the crucial gap between energy generation and energy use, and optimizing the thermal properties of structural materials in order to increase the associated buffer effect. Thermal energy storage can be obtained with three basic kinds of applications: sensible, latent and thermochemical energy storage.

In sensible heat storage, the energy is stored by raising the temperature of the storage medium. In this view, sensible storage materials need to guarantee at least two main physical characteristics: high specific heat and adequate thermal cycling stability. Latent heat storage, however, relies on the phase change transition of the storage medium, e.g. solid-liquid transformation (PCMs phase change materials), to store heat in the form of latent heat of fusion. These materials are characterized by a high-energy storage density and can be considered as very promising for passive building applications. Lastly, thermochemical energy storage is achieved by storing a high amount of energy with using a reversible chemical reaction [272–274].

In this context, concrete, which is a relatively inexpensive material with a high-thermal mass, very easy to handle and cast, can be considered as a suitable solution for sensible heat storage. In particular, concrete has extensively been studied by the scientific community as a high temperature storage material, and specific compositions have been defined in order to achieve long-term stability and increased storage capacity [275–284]. Different kinds of aggregates have been studied in order to improve the compound temperature resistance, i.e. iron and aluminum oxide, gravel, basalt and sand.

In recent years, the problem of high vapor pressure inside the cementitious matrix heated above 100°C has also been investigated. In order to avoid harmful consequences on the integrity of the concrete block, in fact, the cementitious matrix must provide a suitable steam permeability, thus allowing the vapor to leave the storage block. In this context, Laing *et al.* [275] defined a stable mixture using blast furnace as a cement

binder, gravel and sand with improved temperature resistance as aggregates, and a small amount of PP fibers to ensure permeability. Such mixture enabled to obtain significant thermal properties, i.e. a thermal conductivity of $\sim 1.2\text{ W/mK}$ and a volumetric heat capacity of about $\rho \cdot c_p = 0.64\text{ kWh/m}^3\text{ K}$ in the range between 300 and 400°C , and also an impressive cycling stability, i.e. the examined concrete samples endured more than 14,000 cycles up to 500°C .

In addition to its sensible storage capacity, concrete amorphous composite nature, also gives the possibility of integrating supplementary ingredients within its structure. In this context, the idea of combining the appreciable sensible heat capacity of concrete, with also high heat of fusion of phase change materials has attracted a lot of research interest worldwide [285–295], and has identified concrete as a possible latent storage medium.

Two are the main phase change materials which can be coupled with concrete: organic, mostly paraffin and acids and inorganic PCMs, generally hydrated salt. Although hydrated salts are associated with high volumetric heat storage and good thermal conductivity, their very high volume change and supercooling effect highly limit their effective use in concrete composites.

Paraffins and acids, however, which generally combine a high latent heat storage capacity with low volume change, have been widely and effectively incorporated in concrete composites. A phase change material can be incorporated within the concrete matrix in three different ways: immersion [285, 286], impregnation [287, 289] and direct mixing of encapsulated PCM [292–294].

The immersion technique is probably the easiest and normally takes several hours. It consists in the direct immersion of a porous concrete in a container filled with melted PCM. It is directly affected by the absorptivity of such porous concretes and generally associated with serious leakage problems. In order to somehow prevent the melted PCM from ‘flowing out’ of the concrete, silica particles have been used as an effective barrier. This method is not suitable for cast-in-situ structures.

The impregnation technique, however, consists of a three-step procedure: (i) firstly, vacuum is generated in the porous aggregate which will be used to produce concrete, (ii) secondly, the aggregate is soaked in the liquid PCM and (iii) finally, the concrete mixture is produced with using the pre-treated PCM-saturated aggregate. Leakage problems can be associated with this method as well, but in this case, PCM flows inside the composite, causing the reduction of its mechanical properties. The last process which can be used to produce a PCM-concrete composite is the direct mixing of encapsulated PCMs within the cementitious matrix. In this case, the PCM needs to be previously encapsulated in chemically stable container. To this aim, mostly polymers but also steel spheres have been used (Figure 4). In this case, the use of the encapsulation process prevents from leakage problems, unless the composite is subjected to high mechanical loads, which may lead the capsule to premature breakage.

From a thermal point of view, the effect PCM inclusion in concrete is deeply influenced by the kind and amount of PCMs

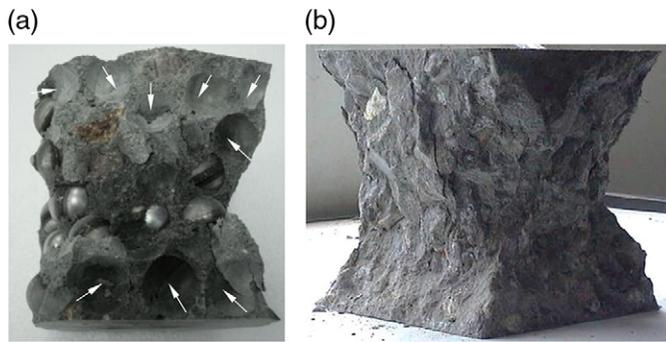


Figure 4. Typical failure patterns of concrete cube (a) steel-encapsulated PCM-concrete and (b) normal weight aggregate concrete [294].

used in the specific mixture. Nevertheless, it possible to find some common trends in the different research contributions. The inclusion of PCMs in concrete generally yields a significant improvement in the thermal performance of concrete: the thermal conductivity of the composite always decreases and the amount of such a decrease is always proportional to the percentage of PCM included in the mixture [291,292], as for the specific heat capacity, it has been found to increase consistently with the PCM percentage [291,292].

5 CONCLUSIONS

Concrete is by far the most widespread construction material worldwide, whereby its diffusion derives from several reasons, including economic competitiveness, reliability, architectural versatility and good mechanical properties. Despite the inherent traditional character of concrete as a structural material, concrete technology is facing a continuous improvement and applied material research in this field is very active and scientifically lively. Owing to its large use, and to its related production processes, concrete contributes to ~7% of total emissions of carbon dioxide in the world. For this reason, most of the research devoted to innovations in concrete and cement-based materials has been recently focused on reducing the environmental impact of cementitious constructions.

This work is a review of recent research trends of low-carbon technologies in concrete, in the perspective of reducing the carbon footprint of constructions. As discussed in the article, three main areas about low-carbon concrete are identified which pursue this overarching objective:

- (i) concretes with enhanced physical and mechanical properties,
- (ii) concretes for reducing the use of raw materials,
- (iii) concretes for energy efficiency.

The first research area includes new concretes which combine the reduction of high-energy components with the enhancement and the rising of peculiar properties. HSCs can allow a strong reduction of structural volumes, while high durable and

fire resistant concretes allow an increase of the service life of the structural elements, generating more environmentally sustainable constructions. Furthermore, new LWCs show an environmental-friendly behavior owing to the reduction of dead loads resulting in smaller structural volumes, as well as to their easier industrialization and casting. Additional improvements can be achieved by the inclusion or addition of innovative particles and fibers within the concrete matrix. Novel concretes with self-healing, photocatalytic and self-sensing properties, realized with the addition of new engineered fillers, represent the opportunity to arrange multifunctional high-technological smart structures with notable sustainable benefits.

The second identified research area regarding low-carbon technologies in concrete concerns environmentally aware production processes that reduce the use of raw materials, by therefore reducing the life-cycle impact of the composite materials and of the whole construction, as well. The first instruments to reduce emissions are the reduction or the replacement of environmental impact materials in the concrete binders and in other components, or the modifications of concrete production methods, using emerging technologies in energy saving. Another important action against greenhouse emissions in the field of concrete industry is represented by the reuse of waste materials within the mixes. This practice allows the reduction of the natural resources' consumption and the disposal of waste materials resulting from different production processes. An innovative way to minimize carbon emission is also the choice of alternative constructive solutions, which result in a reduced energy consumption.

The third analyzed area of research concerns special concretes that are specifically engineered and tailored for energy-efficiency targets. The literature reviewed in this part highlights that some innovative concretes can nowadays be considered as energy-efficient materials, thanks to the wide possibility to integrate cement-based materials with key additives responsible for thermal insulation, thermal capacity enhancement, or optical properties improvements of building envelopes. In particular, the addition of engineered materials and the optimization of the mix designs permit the development of concretes with enhanced energy characteristics, such as special LWCs with high-thermal insulation properties, smart concretes incorporating phase changing materials for enhanced thermal storage capabilities and innovative concretes with improved optical properties and high albedo characteristics.

Overall, this review has shown that concrete can be regarded as a quite promising material for low-carbon applications in buildings, thanks to the recent research developments presented in this work, even if it has been historically considered as an environmentally impacting material. Now, the challenge for this traditional building material is its evolution in a industrialized multifunctional high-performance material with enhanced physical, mechanical, thermal and energy efficiency peculiarities, suitable for applications in the field of sustainable constructions. Possible applications are different and multidisciplinary. They include the implementation of structural and non-structural

elements, structural restoration, flooring, panels, hydraulic and geotechnical elements and infrastructures for smart cities.

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