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Review

The greening of engineered cementitious composites (ECC): A review

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ABSTRACT

Concern over sustainability in the construction industry is growing. Engineered cementitious composites (ECC) have the potential to reduce the carbon and energy footprints of the built environment due to their crack resistance and self-healing properties. Over the last decade, continuous efforts have been made in the development of greener ECC. These efforts can be broadly classified by the use of greener binders, fillers, and fibers. This paper reviews recent progress in the exploration of more environmentally friendly and perhaps even more economical materials, and points to research needed for further enhancing the mechanical or durability properties of ECC. Specifically, the significant contribution of fly ash (FA) as a green binder, and alternatives that address its possible shortage are discussed. The adoption of greener sands (natural or recycled) and fibers (man-made or natural) based on physical, chemical, and mechanical perspectives is evaluated. Further explorations of the ductility of high-volume limestone calcined clay (LCC)-blended ECC, interactions between ground-glass pozzolans (GP) and ceramic powder against alkali-silica reaction (ASR), chemical interaction between recycled ceramic aggregate and cementitious matrix, characteristics of local polyvinyl alcohol (PVA) fibers, and the combined effects of eco-friendly ingredients are recommended.

1. Introduction

Engineered cementitious composites (ECC), since their development, have been known as a class of cementitious materials uniquely possessing superior ductility, strain-hardening properties, and many other advantages over normal concrete. For example, ECC as a family of materials attains tensile strain capacity several hundred times that of normal concrete. Fig. 1 shows the characteristic strain-hardening behavior accompanied by self-controlled micro-cracking of ECC. Such notable functions are bestowed by applying micromechanics theory that attempts to unite multi-scales of mechanical interactions between fiber, matrix, and their interfaces. The goal of ECC design is to suppress the well-known brittleness of cementitious materials, in favor of multiple microcracks under tension [1].

Among traditional ECC compositions [2], ordinary Portland cement (OPC), polyvinyl alcohol (PVA) fiber, and silica sand serve as binder, fiber reinforcement, and fine aggregate, respectively. It has been recommended that these three key ingredients be substituted by greener alternatives because of rising global concerns for more environmental sustainability in the construction industry. High energy intensity and release of high levels of carbon dioxide during the production of cement as well as the carbon intensity of the fine manufactured sand and the synthetic oil coated PVA fiber have attracted much attention from researchers and producers, leading them to engage in developing more eco-friendly ECC through appropriate material selection [1,3].

In addition to the greening in the production phase, the use phase of ECC also strongly influences the sustainability of civil infrastructure due to the reduced maintenance needs associated with the durability of the material [4–7].

In this review of different approaches to the greening of binder, aggregate, or fiber in ECC, relatively recent works (since 2017) are discussed. Further explorations and potential requirements for green and sustainable ECC to be adopted in future field-scale applications are also suggested.

2. ECC with green binder/filler

Supplementary cementitious materials (SCMs) have been recognized as promising ingredients for enhancing the greenness and/or performance of cementitious binders in concrete. Some are intentionally produced, while others are from waste streams from different industrial sites (e.g., coal-fired electric power plants, steel mills, and silica-metal plants). These materials are considered valuable due to their reduced environmental impact compared with OPC [8]. In the case of waste

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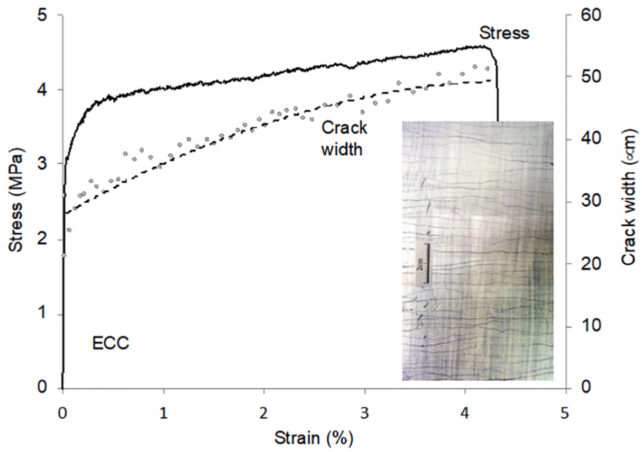

Fig. 1. Typical strain-hardening behavior of ECC, reaching a strain capacity of several % while keeping crack width less than 100 μm [1].

stream materials, their use as SCMs contributes to sustainable practice in other industries beyond cement production and concrete construction.

The current primary SCMs [9] include fly ash (FA), ground-granulated blast-furnace slag (GGBFS) [10–13], silica fume [10,14–17], cement kiln dust [18] and calcined clay (or metakaolin) [19–21] (Table 1). FA is the most frequently adopted SCM in ECC. Table 2 compares the fundamental characteristics of green binders and fillers. Fig. 2 highlights graphically the substantial reduction in embodied carbon of most SCM materials, relative to OPC. (The exception, MgO, is included because of the significant potential for carbon sequestration via carbonation curing, despite its higher embodied carbon compared to OPC). In the following sections, the major contributions of FA to eco-friendlier and more sustainable ECC are described. The potential challenge that FA may face a shortage in the long run is also discussed.

2.1. Benefits of fly ash (FA) in ECC

Produced as a coal combustion residue in thermal power stations [70,71], FA is a well-known substitute for OPC. The advantages of FA include the mitigation of heat release rate, the reduction of CO_2 emissions and lowering of embodied energy [15–17,28], and enhancement of workability. Because of its lower heat of hydration [15], cementitious materials using FA experience less thermal cracking risks, resulting in improvement of long-term durability. Yu et al. [15,17] reported that if OPC were replaced with 80% FA, roughly 70% reduction in total hydration heat, 15% reduction in material production cost along with 70% reduction in CO_2 emissions, and 60% reduction in embodied energy can be achieved for an OPC-based concrete targeting compressive strength of 45 MPa.

Table 3 summarizes the material sustainability indicators in terms of carbon emission, embodied energy, and solid waste generation per m^3 of materials for a conventional concrete and several types of ECCs. ECC with a high volume of fly ash HVFA-ECC (e.g., mass ratio of FA/OPC greater than 1.2) outperformed the typical M45 ECC in embodied energy and CO_2 emissions, while still possessing a decent capacity in terms of tensile strength and tensile strain (i.e., approximately 2.5 to 6.0 MPa in tensile strength and around 2 to 5% tensile strain capacity) [16,24,28]. The higher cost of ECC relative to normal concrete is associated with the cost of fibers, despite typically involving two or fewer percent. Fig. 3 highlights the substantial reduction in embodied carbon of greener versions of ECC relative to the traditional version (M45). In some cases (e.g., versions of HVFA-ECC and PET-PVA-ECC), they are competitive with normal concrete. MgO-ECC with carbonation curing attains a carbon footprint lower than that of normal concrete.

Fly ash has significantly contributed to the greening of ECC. Additional benefits of FA can be found when combined with other additional binder/fillers in ECC mixtures. This will be presented in the next section.

2.2. Combining FA and other additives

SCMs combining FA and other additives have been studied, as a means of further greening ECC while maintaining ductility and durability. Remarkably, FA is not only beneficial as an OPC substitute but also possesses favorable interactions with other eco-friendly materials, as discussed below.

2.2.1. Hollow glass microspheres and FA

Hollow glass microspheres (HGM), a controlled dimension hollow glass material with encapsulated air [62], is considered as an eco-friendly and economical filler in ECC mixtures. HGM effectively lightens the overall material, resulting in lower composite density and inertia [14]. Another advantage of incorporating the spherical and smooth-surfaced HGM in ECC is the improved fresh properties (e.g., workability, flowability, compactability, or dispersion of fiber) [14,62–66] of the composite, which is limited by a low water to cement ratio and the presence of microfibers [1].

Despite the merits of adopting HGM in ECC, it has been reported that their use could impair durability due to alkali-silica reaction (ASR) caused by the chemical reaction between high silica and alkali pore

Table 1
Green binder/filler adopted in ECC.

Type		Green binder/filler	Reference
Binder	OPC-based	Fly ash (FA)	[15–17,22–29]
		Ground-granulated blast-furnace slag (GGBFS) (+FA)	[10–13]
		Silica fume (SF) (+FA)	[10,14–17]
		Rice husk ash (RHA)[a]	[30–34]
		Ground-glass pozzolans (GP) (+FA)	[35–38]
		Solid waste ceramics + FA	[18,39,40]
		Iron ore tailings (IOTs) (smaller particles)	[41]
	Non-OPC-based	Magnesium oxide (MgO) + FA	[42–48]
		Magnesium oxychloride-based (Sorel) cement + FA	[49–51]
		Limestone calcined clay (LCC)[b] (+ FA)	[19–21,52–54]
		Calcium sulfoaluminate cement (CSA cement) (+ FA)	[28,52–61]
Filler		Rice husk ash (RHA)[a]	[30–34]
		Hollow glass microspheres (HGM)	[14,37,62–67]
		Iron ore tailings (IOTs) (larger particles)	[68,69]

[a] RHA serves as both a binder and a filler.
[b] LCC is blended with OPC to be used as limestone calcined clay cement (LC^3).

Table 2
Characteristics of green binders/fillers. OPC is included as a reference material.

Material	Advantages	Disadvantages	Embodied carbon footprint		Embodied energy	
			[kg/kg]	[kg/m^3]	[MJ/kg]	[GJ/m^3]
FA [15–17,22–28,72,73]	Enhances ductility and workability Reduces heat of hydration and shrinkage	Lowers early strengths Possible shortage	0.00–0.01	0–26	0–0.11	0.0–0.3
GGBFS [10–13,74,75]	Increases compressive strength	Lowers early strengths Lowers ductility	0.026–0.14	73–415	1.6	4.5–4.6
SF [10,14–17,75,76]	Increases compressive strength	Lowers ductility	0.014–0.028	31–62	0.036	0.1
RHA [30–34,77,78]	Enhances overall mechanical properties	Uncertain effect on cement hydration Lowers workability	0.157	204–356	10.3–13.3[a]	13.4–30.2[a]
GP [35–38,79,80]	Enhances early strength	Hinders ductility Potential ASR risk	0.0072–0.063	19–190	0.060–0.14	0.15–0.42
Solid waste ceramics [18,39,40]	Enhances ductility		–	–	–	–
IOTs [41,69]	Improves the strain-hardening behavior	Reduces compressive strength	0.026–0.14[b]	73-415[b]	1.6[b]	4.5–4.6[b]
MgO [42–48]	Superior CO_2 sequestration Enhances ductility	Impairs workability	1.4	5040	2.4	8.64
LC^3 [19–21,52–54,81,82]	Refines pore structures	Higher water demand	0.56	1344–1562	4.0	9.6–11.2
CSA [28,52–61,83–85]	Reduces autogenous shrinkage Early strength	Relatively high cost	0.55–0.75	1595–2325	3.6–4.1[c]	10.4–12.7
HGM [14,37,62–67]	Reduces density Improves fresh properties	Increases the risk of alkali-silica reaction	–	–	–	–
OPC [86]	Widely available	Higher carbon and energy intensity	0.84–1.1	2646–3465	4.8–5.5	15.1–17.3

[a] Can be neutral if the calorific value (heat of combustion) of rice husk was considered [78].
[b] Assumed to be the same as those for GGBFS [41].
[c] Assumed to be 25% less than that of OPC [83].

solution [89]. In this case, glass is the source of the high silica content. Expansion and corresponding cracking are caused in ECC, eventually diminishing the composite durability [37,67]. Deteriorations in compressive, tensile, or flexural strength have been also reported [14,62–65,90,91].

While it has been suggested that ASR is passively reduced if the size of glass is relatively small (i.e., less than 0.3 mm [92]) or the surface state is smooth or non-cracked, FA was found to suppress the property deterioration associated with ASR in ECCs containing HGM [14,62,63,65] by diluting the alkalinity of pore solution and refining the pores [67,93]. Fig. 4 shows an example of expansion rate due to ASR in ECC mixtures, indicating that there is an increasing trend in the volume fluctuation with an increase of glass content. That said, the trend was within the range of shrinkage, which was attributed to the presence of FA as well as the small glass particle size [67]. This implies that, even if recycled and granulated glasses are included in ECC where ASR is more likely to occur, the undesirable consequence could be alleviated because of the chemical and physical contribution of FA. The degree of ASR mitigation varies with the types of HGM added [63,65].

Aslani and Wang [63] evaluated the compressive and flexural strength of ECCs at the age of 28 days with three different types of HGM (Table 4). The experimental results suggested that a 60 vol% replacement of FA by one of the three HGM (i.e., Y12000) as the best, within the limited experimental variables. That was because, compared with control ECC that contained only FA, a 3% reduction in compressive strength and a 14% increase in flexural strength were recorded, while achieving a lighter ECC mixture (i.e., the material density from 1,960 to 1,745 kg/m^3). Analysis using scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDS) further clarified the presence of cement particles only around Y12000 HGM, whereas the particles were not confirmed in the other two HGMs and the surface of each remained smooth, implying that the size or surface coating influences its reaction with cement particles.

2.2.2. Magnesium oxide (MgO) and FA

Magnesium oxide (MgO) is a promising candidate requiring less energy than OPC for the calcination process, which involves the heating of inorganic materials to remove volatile components [94]. MgO binds with other ingredients (e.g., fine aggregate or synthetic fiber) in ECC mixture when mixed with a concentrated solution of magnesium chloride ($MgCl_2$), resulting in magnesium oxychloride cement (MOC) [49–51,95,96]. MgO can serve as a binder in another way through mineral carbonation [42], where CO_2 can be positively utilized to activate the binding capability. In this case, incorporation of FA, as well as MgO in ECC, was found effective to further secure the environmental and economic advantages, while considering the mechanical and durability properties [42,43,47,48].

Wu et al. [42] showed that ECC with 100% OPC mass replacement by 50% MgO and 50% FA was better than that with the cement substituted by 70% MgO and 30% FA in terms of environmental and economic impacts, as presented in the second half of Table 3. While MgO itself has higher embodied CO_2 than OPC (i.e., 1.4 kg CO_2/kg vs. 0.85 kg CO_2/kg [97]), the net CO_2 of MgO-based ECC can be lowered compared with OPC-based ECC due to CO_2 sequestration through carbonation curing as mentioned before.

Despite acceptable strain-hardening behavior regardless of the duration of carbonation curing (up to 7 days), the study [42] cautioned a negative correlation between the curing duration and tensile strain enhancement. That is, a higher degree of CO_2 uptake could cause a loss in tensile ductility, attributed to excessive fiber/matrix interfacial bonding that leads to premature fiber rupture in the crack bridging process. This negative tendency was less pronounced in the case of 50% MgO and 50% FA for the binder composition than that of 70% and 30%, respectively.

The use of MgO and additional carbonation curing were found to be effective in reducing the crack width and increasing the number of cracks [42,43,47,48], implying that further improvement of durability could be achieved. Wu et al. [43] reported that 6% weight replacement of OPC by MgO powder helped to reduce hydraulic conductivity by roughly 55%, compared with the control plain ECC, in which both types of specimens had a high FA to OPC ratio of more than 1.0. Although the self-healing ability of those MgO-ECCs was also verified [43–46], it is noted that this ability could also be attributed to the contribution from OPC. Further clarification of ECC properties when OPC is completely replaced by MgO and FA in the binder would be helpful.

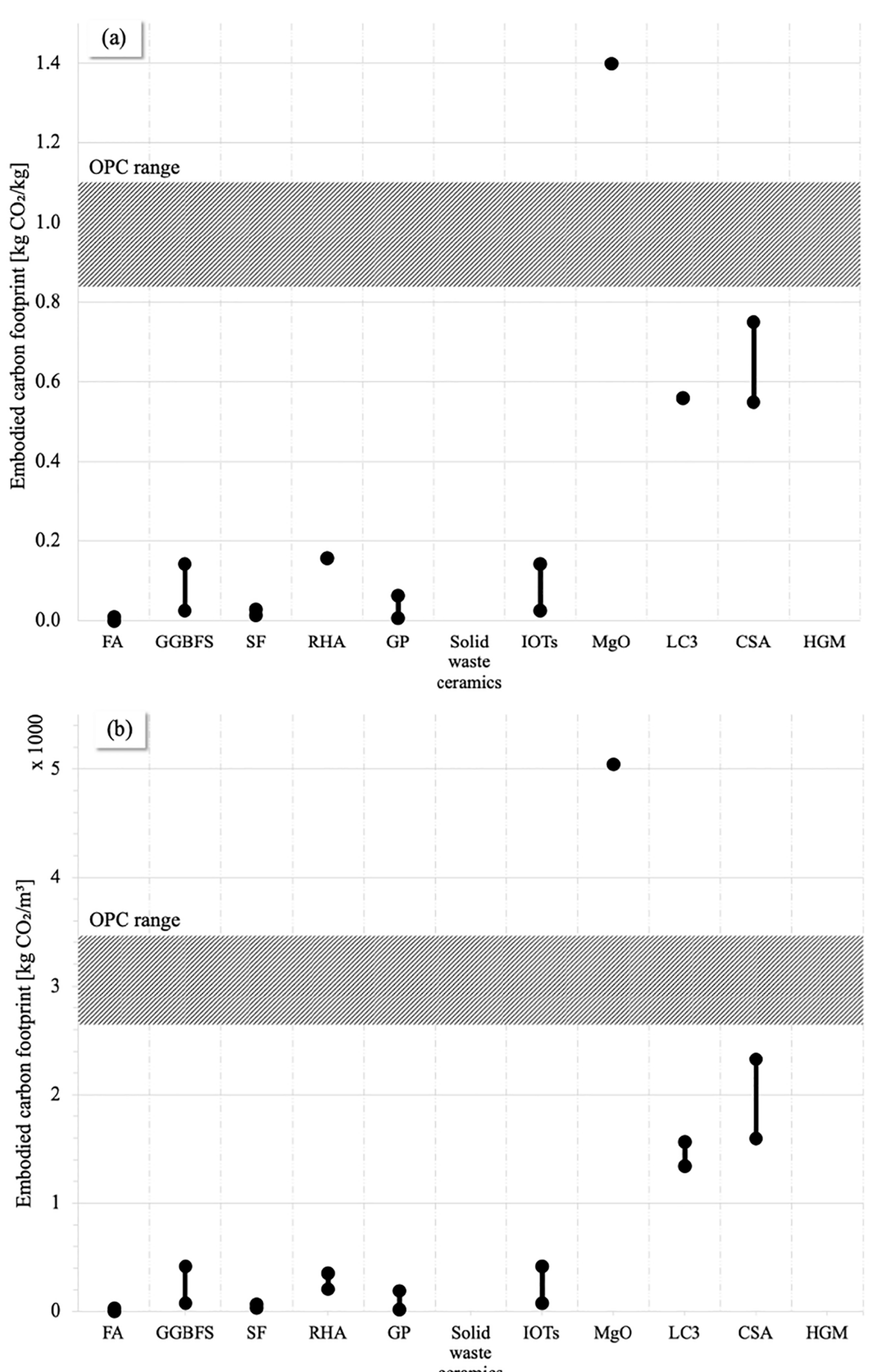

Fig. 2. Embodied carbon footprint of green binders (a) by unit mass and (b) by unit volume.

2.2.3. *Solid waste ceramics and FA*

Solid waste ceramics derived from the manufacturing of tableware ceramics, bathroom ceramics, exterior wall ceramics, and floor tile ceramics [98] raise growing concern for the environment as they are not degradable and are likely to contain harmful substances for land, air, or water resources [39]. Attempts have been made to utilize such waste after fabrication or crushing and grinding [39,40,99,100] as binders or as coarse/fine aggregate in cementitious materials [101–104].

Solid waste ceramic powder (CP) [18,39] and recycled brick powder (RBP) [40,105] have been incorporated into FA-included ECC. The addition of RBP revealed improvement in flexural deflection limit but showed little influence on other mechanical behaviors (e.g., tensile, compressive, flexural strength, and strain-hardening) of ECC. The addition of CP was found to improve flexural deflection and bending toughness, attributed to CP's reactive components for pozzolanic reaction (i.e., more SiO_2 or Al_2O_3 to react with $Ca(OH)_2$), thereby decelerating early hydration and improving the deformation capacity. An important finding of the above studies is that FA plays an important role in ensuring composite ductility in the presence of CP and RBP.

While the re-purposing of solid waste ceramics as green resources in cementitious materials has been demonstrated, the concern of the energy and cost required to grind ceramic waste for use as a binder in green

Table 3
Material sustainability indicators and cost comparison of conventional concrete and various ECCs (adapted from [16,17,29,42,87,88]).

Material	Total embodied energy [GJ / m^3]	CO_2 emission [kg CO_2 / m^3]	Solid waste [ton / m^3]	Cost[c] [USD / m^3]
Conventional concrete	2.46–2.76	260–421	0.20	69–75
Typical ECC (M45)	5.99–6.74	582–629	−0.49	443
HVFA[a]-ECC (FA/OPC[b] = 3.6)	4.35	350.8	−0.84	–
HVFA[a]-ECC (FA/OPC[b] = 4.4)	4.52	279.8	−0.97	429
HVFA[a]-ECC (FA/OPC[b] = 5.6)	3.91	277.0	−0.95	–
ECC (70% MgO – 30% FA)	3.7	451-551[d]	–	308
ECC (50% MgO – 50% FA)	1.9	132-205[d]	–	178

[a] High-volume fly ash.
[b] FA to OPC mass ratio.
[c] Subject to market price fluctuation.
[d] After considering carbon sequestration enabled by carbonation curing.

ECC remains.

2.2.4. *Calcium sulfoaluminate (CSA) cement and FA*

Calcium sulfoaluminate (CSA or Ye'elimite) cement is generally produced by sintering limestone, bauxite, and sometimes industrial waste (e.g., fly ash, slag) at 1200–1250 °C in rotary kilns and by blending with gypsum afterwards. The thermal energy is primarily used for the calcination of limestone [106]. The energy input and CO_2 emissions associated with the production of CSA cement are roughly 25% and 20% less than those for OPC, respectively [59,83]. This is because of the relatively lower calcination temperature (compared with 1,400–1,500°C for OPC) and the carbonate content of its ingredients (e.g., less limestone) [60].

As an alternative binder for ECC, CSA cement is comparable to OPC in terms of mechanical properties [85]. It is further possible to attain reduced shrinkage [28,61] as well as increased early strength [106] due to the presence of ettringite, a sulfate mineral generated by hydration processes (between tricalcium aluminate (C_3A) and calcium sulfate usually as gypsum) [107]. CSA cement can be applied to sprayable or self-stressing ECC where early hardening or exertion of internal pressure is desired [52–54,61].

Partial substitution of CSA cement by FA further enhanced the microstructure and promoted earlier and more ettringite generation [55,56,108]. This is because the nucleation effect of FA is observed [109], and alumina and silica supplied from FA react with monosulfate, a hydration product of CSA without calcium sulfate, to form additional ettringite [58]. Ettringite can cause an expansion in ECC, while it can fill in the pores and enhance compressive strength if it is restrained [57]. Use of up to 15–20% FA in CSA cement was found to be effective in sustaining mechanical (i.e., tensile and flexural) properties under an unconstrained setting [58] and improving carbonation resistance by refining pore structure [56].

Despite the advantages of CSA cement, it should be noted that the availability of bauxite, a primary aluminum-rich source of CSA cement, is limited (e.g., Australia, Guinea, or China) [110]; the energy and cost required to transport CSA can surpass the energy saving in its production [80,100], which hinders its widespread commercial use. The reduction of the aluminum-rich phase in CSA cement produces so-called Belite-Ye'elimite-Ferrite (BYF) cement, which is distinguished from CSA cement and brings an economic benefit due to a direct decrease of the aluminum demand [111,112].

2.3. *Engineered geopolymer composites (EGC)*

FA is one of the most important ingredients in engineered geopolymer composite (EGC), which is distinct from ECC in terms of the mechanism of chemical reaction for hardening [113]. That is because OPC can be completely substituted by FA (or other industrial byproducts such as GGBFS) for OPC as the primary binder [12,23]. While the binder is different, EGC and ECC share a common composite design basis for tensile strain-hardening.

Besides its eco-friendliness, well designed FA-based EGC possesses

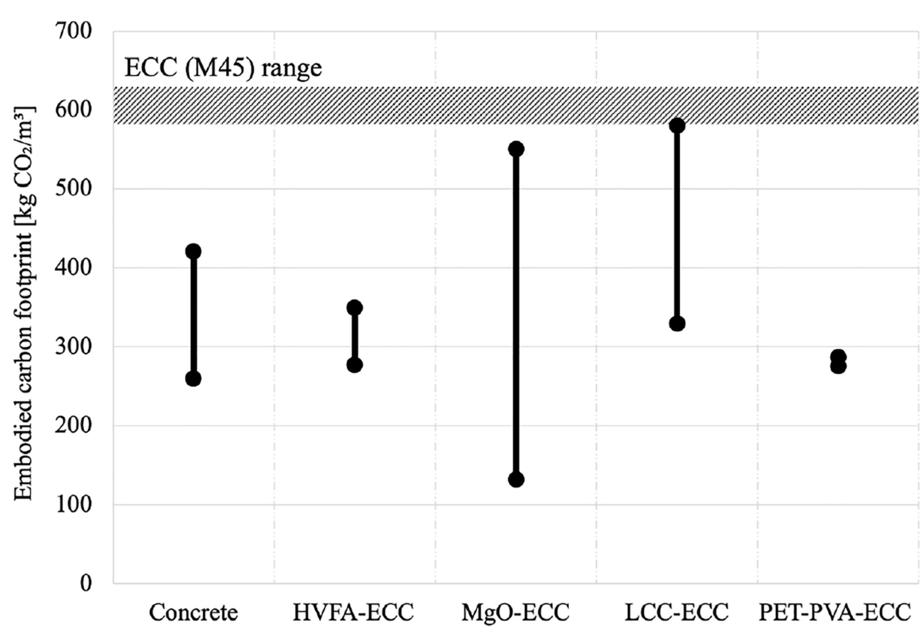

Fig. 3. Embodied carbon emission for conventional concrete and several types of ECCs.

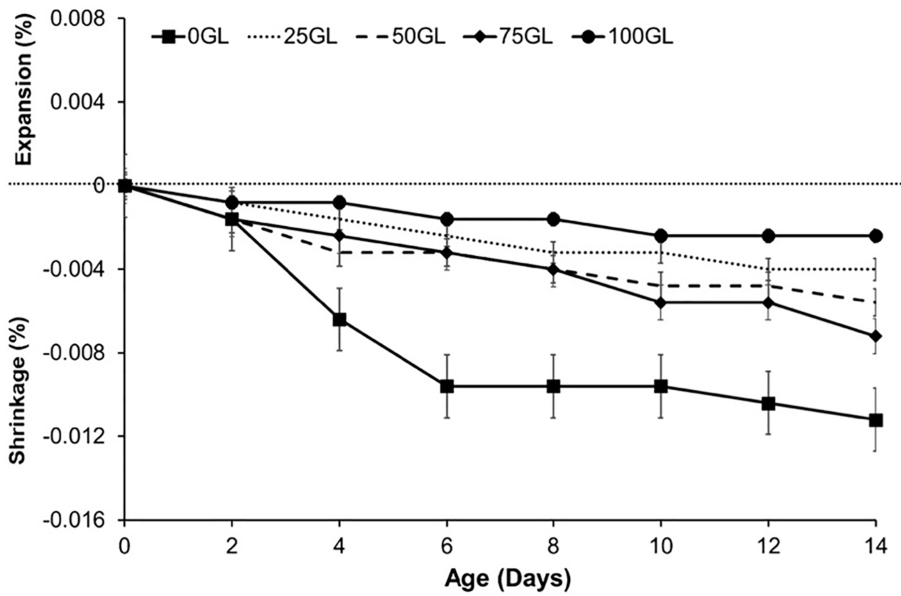

Fig. 4. Expansion over time due to ASR in ECC mixtures [67]. According to ASTM C1260, expansion lower than 0.10% at 14 days after both soaking in sodium hydroxide solution and exposure to 80°C is considered harmless expansion. FA was added to all the five mixture compositions listed in the legend. Note that "#GL" stands for #% glass replacement of ultrafine silica sand. The harmless consequence due to ASR was attributed to the presence of FA and the small glass particle size.

Table 4
Properties of HGMs and strengths of HGM-blended ECC (adapted from [63]).

Product[a]	Density [g/cm^3]	Test pressure[b] [MPa]	Particle size [μm]			Compressive strength[c] [MPa]		Flexural strength[c] [MPa]	
			D_{10}	D_{50}	D_{90}	40%[d]	60%[d]	40%[d]	60%[d]
H25	0.25	5	30	60	120	42	31	7.0	7.7
H40	0.40	28	25	50	95	45	35	6.5	7.0
Y12000	0.60	82	22	48	90	49	45	7.7	8.3

[a] H and Y account for two different surface coatings applied.
[b] represents compressive pressure that each type of HGMs could withstand.
[c] approximate values (28 days) from the figures in the corresponding literature.
[d] #% stands for the volume replacement ratio of FA by HGM.

similar tensile/flexural properties (e.g., strain-hardening behaviors along with decent strength and ductility [25]) and perhaps better durability (e.g., lower drying shrinkage and better resistance to freeze–thaw/wet-dry cycles or acid attack [114–116]), compared with ECC, although the compressive strength is likely lower [25].

EGC production can be more expensive than ECC [117] and a deposit of salt known as efflorescence can be formed on a surface through its hardening process, which is not usually harmful but aesthetically undesirable [118,119]. Detailed studies on EGC can be found in [25–27].

As presented in this section, FA has advantages beyond its greenness as an SCM in the binder of ECC. Those advantages include (a) mitigation of alkali-silica reaction in cases where hollow glass microspheres are present; (b) enhanced eco-friendliness and mechanical properties in magnesium oxide-included ECC; (c) retention of ductility in cases where solid waste ceramic powder coexists with FA; (d) complete replacement of OPC in engineered geopolymer composites.

2.4. Countermeasures to a possible shortage of FA

As presented above, FA has played an important role in partially replacing OPC as a greener binder in ECC. There is, however, a growing concern over the supply of FA due to decreasing reliance on coal combustion as a means of electricity generation across the world, especially in the USA and Canada. As illustrated in Fig. 5, the use of typically inexpensive natural gas or other renewable sources (e.g., wind, geothermal, or solar power) has been increasing and is expected to keep growing, instead of coal combustion secondarily producing FA [35,120]. This trend does not imply an urgent need to shift from FA to other SCMs because more than 40% of total FA produced is not beneficially utilized [121]. Despite such optimism regarding the availability of FA, there are increasing studies on alternative materials to FA, as summarized in the

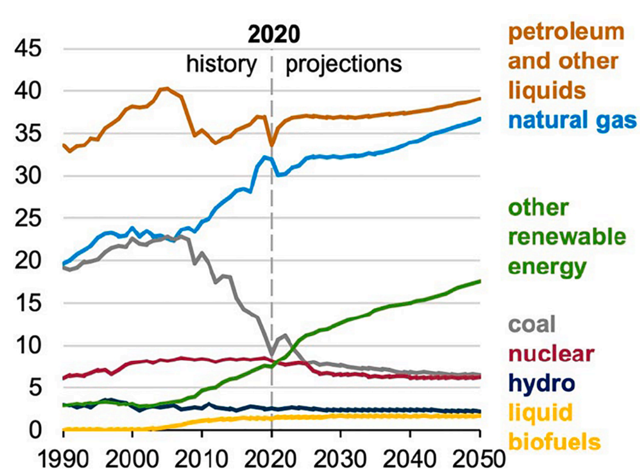

Fig. 5. U.S. energy consumption by fuel annual energy outlook 2021 reference (case quadrillion British thermal units) [120].

section below.

2.4.1. Rice husk ash (RHA)

Rice husk ash (RHA) is an agricultural material obtained by burning rice husk. After combustion, it is traditionally disposed of in water streams or landfills as waste. It has been found that RHA becomes similar to silica fume (SF) in its chemical composition when burnt at a controlled temperature and condition, thereby attracting the attention of researchers [31–34].

Zhang et al. [31] attempted to replace FA with RHA at various ratios of up to 100% blended with a small amount of SF. OPC was partially substituted by RHA as a hybrid binder not containing FA in a study by Costa et al. [32].

Both experimental studies clarified that the inclusion of RHA refined the pore distribution and increased the total pore volume. Specifically, the portion of large capillary pores (e.g., greater than 100 nm) was reduced and that of middle-sized pores (e.g., 50–100 nm) was increased. The modified pore structure was found to improve the compressive strength, tensile strength, and tensile strain capacity, compared with conventional ECC. The enhancement of compressive and tensile strength was attributed to the higher density of RHA-included ECC mixture owing to the packing effect and filler effect of RHA. The finer particle size of ground RHA (compared with FA) reduces physical voids, and its larger surface area creates more agglomeration sites for cement particles [31,122]. The improvement of the tensile strain property was attributed to a narrower crack width and a larger number of cracks, resulting from an increased pseudo strain-hardening (PSH) index [31].

Contradictory findings on the effect of RHA on primary cement hydration have been reported. In [30], decelerated cement hydration was identified by a lower degree of shrinkage. In [31], however, accelerated cement hydration was reported and attributed to the filler effect of RHA. As previously noted, a major distinction between these two studies was their binder compositions in ECC mixtures. FA was replaced by RHA in which a constant amount of OPC and silica fume was present in [31], while OPC was substituted by RHA in the complete absence of FA in [30]. Further research is needed to address the durability concern associated with early-age cracking due to shrinkage and with thermal cracking caused by accelerated cement hydration. Improved understanding of the effects of RHA on the mechanical and durability properties of ECC can make it a viable replacement for FA.

2.4.2. Ground-glass pozzolans (GP)

Ground-glass pozzolans (GP), also known as glass powder, has been used to completely replace FA in ECC binders. GP is obtained by grinding post-consumption glass and is considered environmentally friendly with a carbon footprint of 0.063 kg CO_2/kg [79]. For comparison, FA has a carbon footprint of 0.01 kg CO_2/kg [72] or less, which is nearly negligible. The particle size of GP is controlled from 1 μm to 100 μm equivalent to the fineness of FA [79,123].

GP was found to densify ECC, improving compressive, tensile (the first crack or post crack strength), and flexure strength at early ages [35–38]. This was attributed to the packing effect and filler/nucleation effect. The irregularly-shaped GP provides a larger surface area than the spherically-shaped FA for new pozzolanic calcium silicate hydrate (C-S-H) with a low Calcium/Silica ratio and high alkali or aluminum content, which endows ECC with a denser structure [36,37].

GP may limit tensile ductility by reducing the crack density [35–38]. This reduction was caused by the excessively high packing effect of GP raising the matrix first-cracking strength and the frictional bond τ_0 in fiber and matrix interfaces, which likely induced fiber rupture rather than fiber slipping in a bridged crack. To counter the loss of ductility caused by the inclusion of GP, consideration of nano reinforcement is fruitful within multi-scale modifications [124,125]. Nanofiber (e.g., graphite nanoplatelets or carbon nanofiber), nanocomposites (e.g., graphene oxide (GO), or nanocarbonate whiskers ($CaCO_3$)) could be incorporated into GP-based ECC. Among these, the use of nanoscale cellulose filaments (CF) [35,36] was found to diminish the frictional and chemical bond by modifying the interface between PVA fiber and matrix and restoring the ductility and strain-hardening capability. This positive effect is more pronounced for ECC with a higher volume content of GP.

The durability of GP-based ECC has received less attention. One of the few studies investigated chloride ion penetrations [37] which affects steel corrosion [126]. The degree of rapid chloride penetration was higher in GP-based ECC than that of control (i.e., fly ash to cement ratio of 1.2) ECC specimens at 28 days, because of the higher alkalinity in the pore solution of GP-based ECC [127]. Despite this unfavorable higher chloride diffusion at 28 days, the chloride content appeared to plateau at a similar value at a longer time for ECC with and without GP [37]. This was explained by continued pozzolanic reaction and cement hydration in the presence of GP.

Concerns for ASR deterioration of GP-based ECC have been raised [35–37,39]. It was reported that solid waste ceramic powder (CP) with its superior pozzolanic reactivity similar to FA could mitigate expansion due to ASR. Further clarification is needed in the suppression of ASR by finer glass particle size or through interaction with CP.

2.4.3. Limestone calcined clay (LCC)

According to [128], limestone and kaolinite clay are abundant across the world. Limestone calcined clay (LCC) is produced by blending limestone and calcined clay. Calcined clay, which is a low purity metakaolin, is obtained by calcination of low-grade kaolinite clay at 600–800 °C, whereas OPC requires so-called clinker which is a primary ingredient of OPC and is obtained through calcination at up to 1450 °C, much higher than that for metakaolin. LCC displaces part of OPC to make limestone calcined clay cement (LC^3). The overall energy required to produce LC^3, and corresponding CO_2 emissions are much less (e.g., 22% less energy consumption and 20–35% less CO_2 emissions) than those for OPC [81,82].

While a few studies [20,21] have substituted LCC for OPC, few studies have attempted to take FA as a target to be partially or even completely replaced by LCC, except for [19]. There were some distinctions between those studies in terms of mechanical properties or environmental impacts, which are briefly summarized in Table 5.

First, the relatively low tensile strain capacity of ECC with high LCC content (i.e., 70% or 80% of total binder [19]) should be highlighted, whereas relatively high compressive and tensile strengths were recorded. This could be attributed to a denser matrix due to the high compactness of LCC [20] and loss of contribution of fly ash that indirectly enhances ductility by improving fiber dispersion [31]. Second, the trend of tensile strain capacity varying with different water to binder ratios in LCC-blended ECCs was also different between [19] and [21].

The authors [19] reported the tensile ductility improved along with increased water to binder (i.e., OPC + FA + LCC) ratio, whereas the opposite tendency was observed (i.e., the improved tensile performance was accompanied by the ratio decreased) in [21]. One plausible explanation of this apparent contradiction comes from the different fibers adopted, which affect the interfacial bonding property between fiber and the LCC-blended matrix. According to [18] where hydrophilic PVA fiber was used, an increase in the water to binder ratio was found to cause the porosity to increase and the contact surface area between the PVA fiber and matrix to decrease. This microstructural change resulted in a lower interfacial frictional bond so that more fiber slippage could occur for larger tensile strain capacity, when interfacial chemical bonding was inherently high. In contrast, PP fiber is hydrophobic with weak chemical and frictional bond. The increased water content weakens the interfacial bonds, thereby lowering tensile strain capacity [21]. Further exploration of the interaction between an LCC-included matrix and fiber type is recommended.

For the environmental and economic impacts, LCC-included ECC is overall superior to OPC-based ECC. Substantial reduction in material sustainability indicators in LCC-blended ECCs over OPC-based ECC [20,21] is achieved by the replacement of OPC by LCC, the absence of

Table 5
Comparison of OPC-based ECC and LCC-blended ECC (weight ratio in mixture composition).

	OPC-based ECC		LCC-blended ECC				
Reference	[20]	[21]	[20]		[21]	[19]	
OPC[a]	1	1	0.55	0.55	0.55	0.96	0.64
Fly ash (FA)	2.20	2.20	2.20	2.20	2.20	0	0
LCC[b]	0	0	0.45[c1]	0.45[c2]	0.45	2.24	2.56
Water	0.79	0.95	0.95	0.95	0.95	0.95	0.95
Silica sand	1.16	0	1.16	1.16	0	1.33	1.33
H or S[d]	0.013(H)	0.004(S)	0.022(H)	0.022(H)	0.008(S)	0.020(S)	0.023(S)
2 vol% fiber[e]	PVA	PP	PVA	PVA	PP	PVA	PVA
Compression strength [MPa]	55.3	28.0	32.1	31.3	20.0	64.5	43.6
Tensile strength [MPa]	6.5	3.4	4.2	4.2	2.5	5.19	4.65
Tensile strain capacity [%]	4.2	3.7	6.5	5.1	6.0	0.63	0.57
Embodied energy [GJ/m^3]	9.2	4.1	7.6[f]		3.5	7.5	7.3
CO_2 emission [kg CO_2/m^3]	765	480	473[f]		330	580	495
Cost [USD/m^3]	780	170	685[f]		170	–	–

[a] ordinary Portland cement.
[b] limestone calcined clay.
[c1] limestone particle size was 3 μm.
[c2] limestone particle size was 12 μm, with the same metakaolin as in c1.
[d] H: high-range water-reducing admixture, S: superplasticizer.
[e] PVA: polyvinyl alcohol, PP: polypropylene.
[f] difference of particle size in limestone was negligible.

silica sand, and the use of PP fiber replacing PVA fiber (Table 5).

In summary, there are several alternatives to FA that may be suitable for counteracting the potential shortage of FA supply in the future. Some researchers have attempted to completely replace FA with rice husk ash or limestone calcined clay cement, while others have explored ground-glass pozzolans. There are, however, still obstacles to be cleared before practical use in the field for ECC containing other supplementary cementitious materials in replacement of FA, which include ductility enhancement of high volume LCC-based ECC or assessment of alkali-silica reaction in ECC mixtures containing fine GP. Future research on these kinds of challenges is recommended.

3. ECC with green aggregate

Manufactured silica sand traditionally adopted in ECC incurs economic and environmental costs [1]. Attempts to substitute manufactured sand can be divided into two major categories: natural or recycled sand. Their advantages and disadvantages, together with embodied carbon and energy are summarized in Table 6. Fig. 6 highlights the carbon footprints of green aggregate. Except for crumb rubber and glass beads, a significant reduction relative to manufactured sand can be attained. From a sustainability point of view, however, it is the energy and emissions associated with the transport of heavy sand that is of importance. Hence green aggregates often imply locally available sand. The adoption of industrial waste stream material avoids the economic and environmental penalties of landfilling. ECC mechanical and durability properties associated with each type of green aggregate are discussed below along with Table 6 and Fig. 6.

3.1. Natural sand

One of the most efficient ways to reduce the economic and environmental impacts involved with the production and transport of sand is to prioritize locally available materials for massive infrastructure projects [134]. Specifically for marine or coastal constructions, sea sand can be attractive. Use of sea sand and seawater for normal ECC (containing PVA fiber and FA), was found to slightly decrease tensile strain and tensile strength but promote compressive strength and setting time [135,136,145]. Alternatively, river-sand (RS) is economical in comparison to ultrafine silica sand (USS) [133].

Apart from river or sea sand, desert (or dune) sand has been investigated [13,129–131,146] for replacing manufactured sand in ECC. Increased substitution rate, however, was found responsible for diminished composite elastic modulus, tensile/compressive strength, and tensile strain capacity [129,131].

Size effect. Sand is typically classified into three groups according to its particle size [134]: coarse (<4.75 mm), medium (<2.36 mm), and fine (<1.18 mm) sand [133,134,136,145,147]. Within the sand size range studied, the tensile strain capacity is marginally affected (Fig. 7). That said, finer natural sand likely leads to greater compressive/tensile

Table 6
Characteristics of green aggregate. Manufactured sand is included as a reference material.

Material	Advantages	Disadvantages	Embodied carbon footprint		Embodied energy	
			[g/kg]	[kg/m^3]	[kJ/kg]	[MJ/m^3]
Dune [129–132]	Economical	Impairs ductility and strengths	2.4	6.36	31.4	83.2
River [132–134]	Economical	Reduces compressive strength	2.4	6.26	31.4	82.0
Sea [132,135–138]	Increases compressive strength Promote setting time	Decreases tensile strength and capacity	2.4	6.50	31.4	85.1
Crumb rubber[a] [139–141]	Increases tensile capacity and durability	Decreases strengths	200	220	4000	4400
Glass beads[a] [67,73,142]	Improves strengths and ductility		50	120–140	760	1824–2128
Recycled concrete [132,143,144]	Economical Reduces drying shrinkage	Decreases strengths and ductility Reduces durability	1.2	2.88	16	38.4
Manufactured sand [20,21]	Widely studied Consistent quality	Entails energy and cost to process	23.3–33.0	60.1–85.8	67–226	174–588

[a] Crumb rubber and glass beads derive from waste streams from the tire and glass bottle industries.

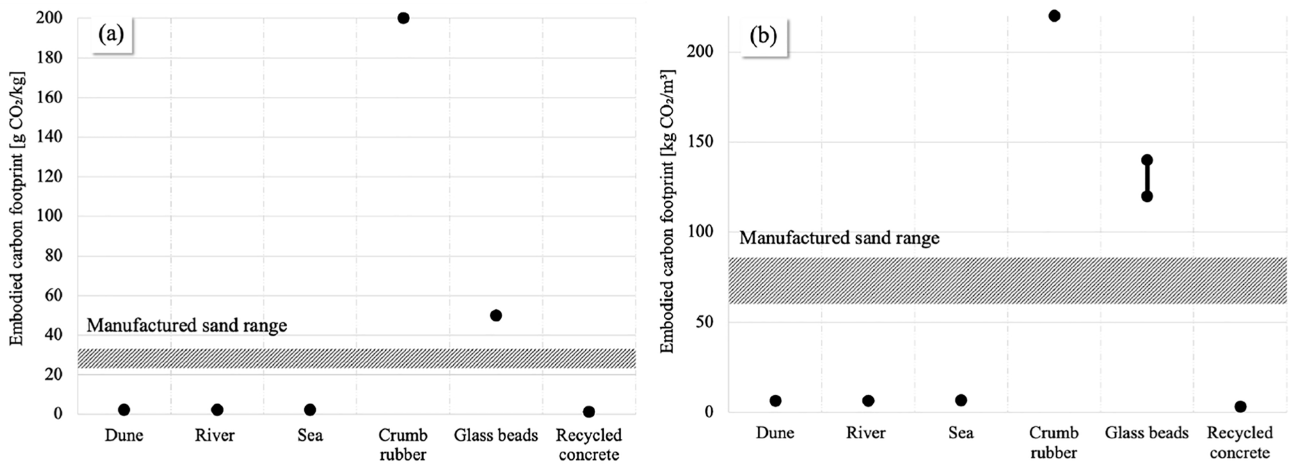

Fig. 6. Embodied carbon footprints of green aggregates (a) by unit mass and (b) by unit volume. Note that crumb rubber and glass beads recycled from waste tires and glass can be still considered environmentally friendly because of the environmental risks caused by their disposal.

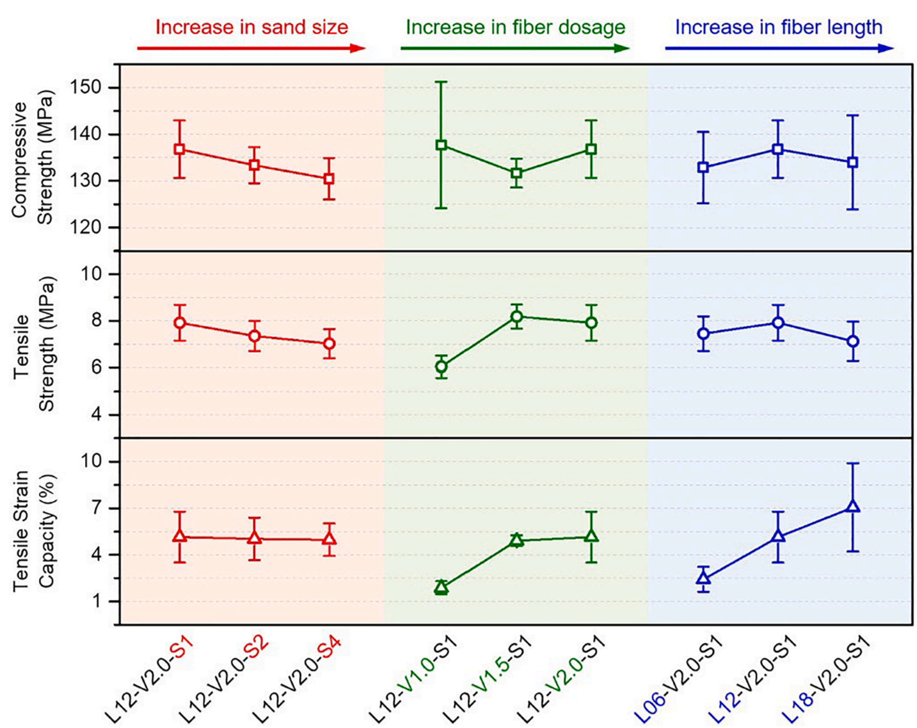

Fig. 7. Summary of compressive strength and tensile properties of sea sand-ECC [145].

strength as well as an enhanced pseudo strain-hardening (PSH) index.

Sand content — sand to binder ratio. As far as river sand, or desert sand as its alternative, is concerned, sand to binder ratio was found to be critical to tensile properties [129,131,133,144], whereas it was not greatly significant for compressive strength or flexural properties (i.e., water to binder ratio plays a primary role in governing compressive strength while fly ash content/fiber volume governs flexural properties) [129,131,133,134,147]. Li et al. [148] revealed that excessive sand relative to binder content leads to a loss of tensile strain capacity and complementary energy, and an increase in the first crack strength accompanied by an increase in crack tip toughness.

The effects of the sea-sand to binder ratio on ECC composite properties, in contrast to the river sand to water ratio presented above, remain to be elaborated, even though they might also be expected to have similar trends.

Morphological impacts. The influence of morphological parameters (i.e., roundness or sphericity) of natural sand on mechanical properties was examined in [13,149]. A strong correlation (coefficient of determination $r^2 = 0.75$–0.89) between these morphological parameters and ECC's mechanical properties was found. The authors discovered that lower roundness and sphericity positively contributed to several aspects, including (a) higher compressive strength due to higher interfacial bonding between sand and matrix; (b) higher tensile strain capacity because of better fiber dispersion uniformity that leads to narrower crack width and more microcracks; and (c) more pronounced strain-hardening behavior caused by a stronger interfacial frictional bond between fiber and matrix that made it possible to have larger complementary energy. It was also noted that the use of sand with low roundness and sphericity required a relatively high dose of superplasticizer to maintain sufficient workability for mixing and casting.

3.2. Recycled aggregate

Apart from natural aggregates, recycled aggregate offers a plausible green alternative. Construction and demolition (C&D) debris is a type of waste that is not included in municipal solid waste and includes concrete, asphalt concrete, steel, wood products, drywall and plaster, brick and clay tile, and asphalt shingles [150]. In 2018, C&D debris of 540 megatons was produced and almost 25% of it was disposed of in landfills in the United States, according to the U.S. Environmental and Protection Agency [150]. There has been a growing interest in re-purposing that landfill waste by using it as recycled industrial aggregate [151–153], eventually aiming at a cleaner and more economical substitution for USS (e.g., recycled concrete aggregate costs 11 times less than USS [154]).

Size effect. The size effect of recycled concrete aggregate (RCA) is more complex than that of natural sand [143,144]. It was found that the compressive, tensile (strain capacity, strain-hardening behavior), and flexural properties of ECC were affected by RCA size. Since RCA is a cementitious material incorporated into ECC as a green aggregate, such aggregate has a self-cementing property and creates additional so-called interfacial transition zones (ITZs), which are usually created by physical or electrochemical (i.e., physicochemical) interactions between the aggregate and the cement matrix [155]. Those interfacial zones caused by the recycled cementitious material are older and weaker than those in a fresh cement matrix, eventually greatly affecting actual mechanical or durability properties originating at the old and new ITZs [107,156]. Finer RCA creates more old ITZs due to its high surface area, but also it is more likely to have its unreacted cement particles exposed to water to further improve the ITZs. This trade-off should be met halfway for optimization (i.e., neither too fine nor too coarse) and this was clarified in [144]. As an example, results of compressive strength and flexural tensile strain capacity varying with RCA/recycled concrete fine (RCF) size are presented in Fig. 8.

Sand content — sand to binder ratio. Sand to binder ratio for recycled concrete aggregate (RCA) was of great importance not only for tensile properties but also for compressive and flexural properties (i.e., the more RCA used, the lower the compressive/flexural strength and strain capacity) simply due to the greater likelihood of activating the old ITZs [144]. It was also revealed that durability properties (chloride ion penetration, permeable voids, and water absorption) were degraded with increased content of RCA, whereas drying shrinkage was reduced due to the internal curing effect of RCA [144,154].

An increase in glass beads aggregate content, in contrast, improved the aforementioned properties as well as permeability properties and alkali-silica reaction resistivity, owing to its physical characteristics (small, smooth, and spherical) and pozzolanic reaction over time [67,142].

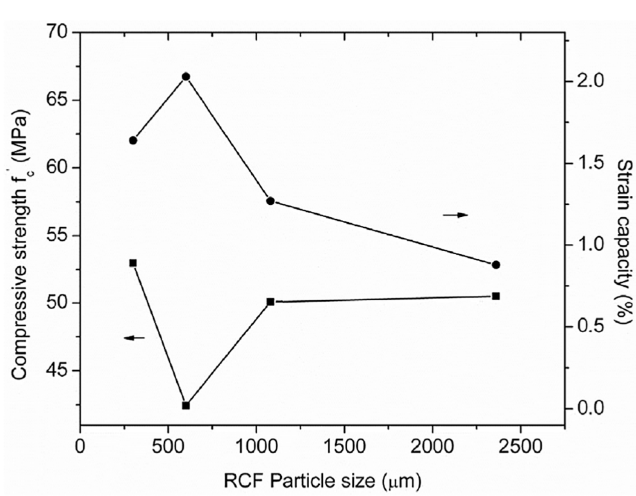

Fig. 8. Compressive strength and flexural tensile strain capacity of recycled concrete fine (RCF)-ECC versus RCF size [144].

An increase in the amount of crumb rubber was found responsible for decreased strength-related mechanical properties (i.e., compressive, tensile, and flexural strengths) and increased drying shrinkage due to its low stiffness which leads to lower restraint within the matrix. In contrast to such negative effects, increased tensile strain capacity and improved durability properties (i.e., porosity, water absorption, and chloride penetration) were also reported, resulting from its low water absorption and pore refinement [140]. The source of enhanced ductility and tight crack width for ECC with crumb rubber remains to be researched.

In summary, there is an increasing amount of literature on the development of green ECC with greener aggregates, based on the physical parameters (e.g., size effect or morphological perspectives) and partly on interfacial reactions between the aggregate and the cement matrix, especially for recycled aggregate. To precisely explain their impacts on ECC composite properties, further study on the interfacial interactions within the matrix, beyond the physical characteristics per se, is recommended. In addition, the role of aggregates on matrix fracture toughness which impacts the stress level to initiate microcracks, and the energy consumed during microcrack propagation warrants additional studies. In some cases, such as crumb rubber, the aggregate may act as artificial flaws affecting the microcrack density and composite tensile ductility. These considerations suggest room for future investigations in ECC composite optimization for material greenness and mechanical and durability performance.

4. ECC with green fibers

Polyvinyl alcohol (PVA) is a synthetic polymer that has received considerable attention in a wide range of applications because of its excellent mechanical properties, thermal stability, and chemical resistance [157]. Further, the fiber diameter (about 40 μm) is small enough to enhance fiber/matrix interfacial surface area critical for composite ductility in the hardened state but large enough to enable good workability in the fresh state. PVA fiber was first commercialized in 1950, with Kuraray Co., Ltd (Japan) as the worldwide producer [158]. The use of PVA fiber imported from Japan in other countries, however, is expensive, which can constitute roughly 50–90% of the total cost needed to produce conventional ECC [24,133]. Furthermore, PVA possesses a relatively high embodied carbon and energy footprint as it is derived from vinyl acetate refined from fossil fuels [3].

In this section, attempts made to address the issues associated with PVA fiber are reviewed, and successful outcomes and corresponding obstacles are presented. Table 7 summarizes the advantages and disadvantages of various alternative fibers that have been adopted in the production of ECC, and their embodied carbon and energies. Fig. 9 highlights the carbon footprints of these fibers relative to that of PVA fiber.

4.1. Modification in PVA fiber — Domestic or unoiled PVA fiber

The PVA fiber (designated REC15) designed for ECC has an oil coating that intentionally reduces the interfacial frictional/chemical bonds between the fiber and cement matrix to induce controlled fiber-slippage for superior ductility and strain-hardening behavior of ECC [173].

For greener ECC, the focus in this paper, some studies [10,24,173,174] have attempted to use domestically produced unoiled PVA fiber. The domestic PVA fiber is effective for reducing the energy involved in transportation and is four to eight times less in cost than that of conventional (i.e., imported and oil-coated) PVA fiber [24,162].

Wang et al. [10] investigated ECC reinforced with unoiled PVA fiber locally produced in China. The authors confirmed reduced composite tensile strain capacity but retained compressive and flexural strength. Possible countermeasures may be to increase the water to cement ratio

Table 7
Characteristics of green fibers. PVA fiber is included as a reference material.

Material	Advantages	Disadvantages	Embodied carbon footprint		Embodied energy	
			[kg/kg]	[ton/m^3]	[MJ/kg]	[GJ/m^3]
PP [159–161]	More locally available, lower cost	Impairs fiber dispersion (High aspect ratio)	2.0–3.1	1.82–2.82	75–115	68.3–105
PE [162–164]	Superior tensile strength	High cost with decreasing trend Larger crack width	2	1.94	73–116	70.8–113
Basalt [159,165,166]	Thermally resistant Enable tighter crack width	Lowers tensile ductility Premature rupture	2.6	6.97–7.28	18	48.2–50.4
Glass [159,160,167,168]	Improves flexural toughness and ductility	Potential premature rupture	0.16	0.416	13–32	34–83
Plant [159,169,170]	Renewable Low cost	Poor durability in cement	–	–	–	–
PET [171,172]	Recycled from the daily plastic products	Compromises tensile strength and strain capacity	0.81–3.4	1.11–4.66	39	57
PVA (reference fiber) [142,173]	Extensively researched	High carbon and energy intensity	1.7–3.6	2.21–4.68	101–106	130–138

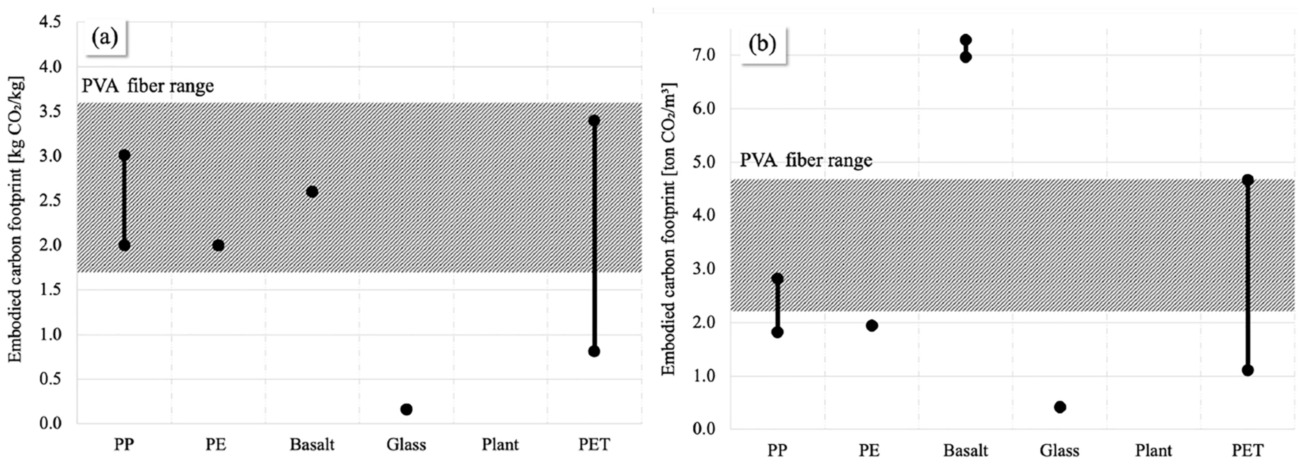

Fig. 9. Carbon footprint of green fibers (a) by unit mass and (b) by unit volume.

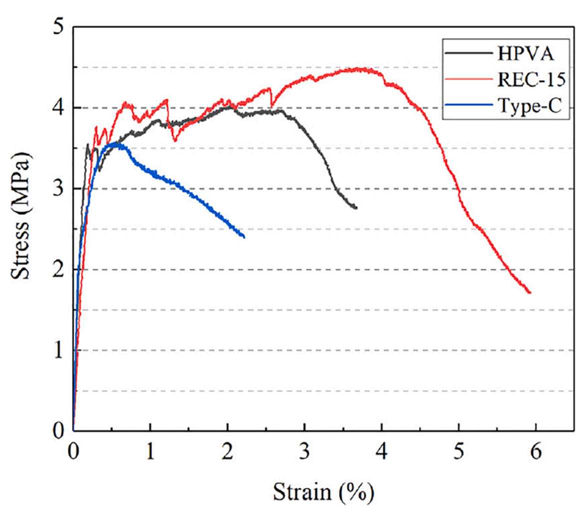

Fig. 10. Typical tensile stress–strain curves. The two cost-effective oil-coated PVA fibers are denoted as Type-C and HPVA, while REC-15 refers to PVA fiber by Kuraray (Adapted from [173]).

or fly ash replacement (of cement) ratio [162]. Despite the expected reduced compressive strength [173], the resulting ECC may be suitable for certain applications. Other attempts involving locally produced oil-coated PVA fiber resulted in ECCs with competitive tensile properties [24,175] and others with relatively low tensile ductility [173,174] (Fig. 10).

While the database for ECCs produced with REC15-PVA fiber is well established, other locally sourced PVA fibers may still be adopted for successful development of greener ECC provided adjustments in binder be made to accommodate the different fiber properties and fiber/matrix interface properties.

4.2. *Adoption of green fibers*

From the environmental and economic points of view, some man-made and natural fibers have the potential to replace conventional PVA fiber. These include polypropylene (PP) fiber, polyethylene (PE) fiber, basalt fiber (BF), glass fiber (GF), and plant fiber, which shall be introduced in this section. Other fibers (e.g., carbon, steel, nylon, acrylic, aramid, animal, etc.) were excluded from the candidates because of the lack of at least one important factor for improving sustainability in ECC, as comprehensively discussed in [159].

4.2.1. *Polypropylene (PP) fiber*

Polypropylene (PP) fiber is cheaper and less energy-intensive than coated PVA fiber as well as more domestically accessible in many countries where PVA fiber is imported from Japan [134]. This alternative fiber has generated ECCs with comparable or higher tensile ductility or durability [159] of PVA-ECC. Some PP fibers also require surface treatment over concern in aging, relatively low chemical bonding (due to its hydrophobicity) [176], and to improve fiber dispersion during mixing caused by its high aspect ratio (i.e., length to diameter ratio) [161]. To enhance their tensile strength, PP fibers have high draw ratios resulting in a lower diameter (e.g., 12 μm) compared to that of PVA fibers (~40 μm). This smaller diameter enhances composite tensile properties by having a larger fiber/matrix contact surface area but also worsens workability and fiber dispersion uniformity.

PP fiber is establishing itself as a greener alternative to PVA fiber [159]. Extensive research in ECC combining PP and green binders (see Section 2 on ECC with Green Binder/Filler) can be found in the literature. These include investigations into the durability properties of RHA-included ECC [30]; flexural ductility and crack-control ability of waste river sand-blended ECC [134,147]; mechanical and self-healing behavior of LCC-mixed ECC [21]; viability of sprayable CSA cement-LCC-blended ECC [52]; and self-stressing CSA cement-mixed ECC [53].

4.2.2. High modulus Polyethylene (PE) fiber

Polyethylene (PE) fiber possesses higher tensile strength (tenacity), higher Young's modulus, lower density, but slightly higher embodied energy and CO_2 (i.e., per unit mass) than those of PVA fiber [159]. PE fiber endows ECC with the most outstanding technical performance capabilities beyond PVA fiber by achieving high tensile/compressive strength and strain capacity [163,177]. To fully utilize its extraordinary tensile strength, surface treatments using ozone [178], silane coupling agents [179], or graphene oxide [180] have been exploited to enhance interfacial bonding limited by PE's hydrophobicity.

One obstacle to the broader application of PE fiber is its much higher cost compared with PVA; about eight times more than PVA fiber [162] and is thereby not practical for large-volume applications. The outstanding composite properties of PE-ECC, however, may justify its use in relatively small-volume applications in which both high composite strength and ductility are required [135,159]. In recent years, newer lower-cost PE fibers have entered the market and are making inroads into larger volume construction applications.

Recent studies have reported the properties of PE-ECC with green binders such as magnesium oxychloride cement [49], rice husk ash [31], recycled fine powder [105], and waste cement kiln dust [18].

4.2.3. Basalt fiber (BF)

Basalt fiber (BF), an inorganic material produced by melting basalt at high temperatures (approximately 1200 °C to 1500 °C), has attracted attention as a high-temperature resistant, relatively inexpensive, chemically stable, and eco-friendly alternative to PVA fiber [159,166]. The relatively high embodied carbon of BF when measured on a unit volume basis (Fig. 9(b)) is due to the higher density of this mineral fiber compared especially to low density synthetic fibers. This implies a carbon footprint penalty to BF as fiber reinforcement performance in a composite is based on volume fraction rather than weight fraction of its ingredients. Xu et al. [165] reported tensile properties of BF-ECC. While tensile strain hardening was achieved, the tensile ductility was limited to less than 1% (Fig. 11). A major advantage of BF-ECC is that the microcrack width is extremely tight, typically below 10 μm. This tight crack is associated with the high stiffness of basalt fibers and its strong bond to the cementitious matrix.

4.2.4. Glass fiber (GF)

Glass fiber (GF), mostly derived from silicate glasses, has lower material sustainability indicators when compared with PVA fiber. For example, the embodied energy and CO_2 emissions per volume of GF are roughly two and five times less than those of PVA fiber [159,160]. GF is prone to corrode or break in high alkali environments; however, mineral admixture (e.g., fly ash, silica fume, or slag) can mitigate the high alkalinity generated by cement hydration, leading to further greening in ECC [167]. Alkali-resistant (AR) glass fiber has been developed [181]. However, durability concern appears to remain [182].

The flexural toughness and ductility [168,183] as well as modulus of rupture (MOR) [184–186], slightly increased with increasing GF content. The MOR of GF-ECC increased with time as a result of increased mortar matrix strength and fiber/matrix bond strength [167].

Other mechanical or durability properties of GF-ECC have yet to be explored. One study did evaluate [187] the high-temperature resistance of ECCs reinforced by various fibers, including GF, PVA, PP, and carbon fiber, and revealed that PVA fiber-ECC performed better than any other type of ECC.

4.2.5. Plant fibers

As a family of natural fibers, plant fibers from the agricultural sector are considered sustainable since they are biodegradable, renewable, and thereby less carbon/cost-intensive than most man-made fibers [159]. The mechanical properties of such natural fibers, which vary due to their various chemical compositions and microstructures, are substantially below those of PVA fiber, even after some fiber processing (e.g., shaping or heat/seawater treatment) [169,188,189]. Some plant fibers, such as Curauá, have reported strength that approaches those of PP fiber [169]. While tensile strain-hardening has been demonstrated, the tensile strength and ductility of plant fiber based ECC remain limited. Fig. 12

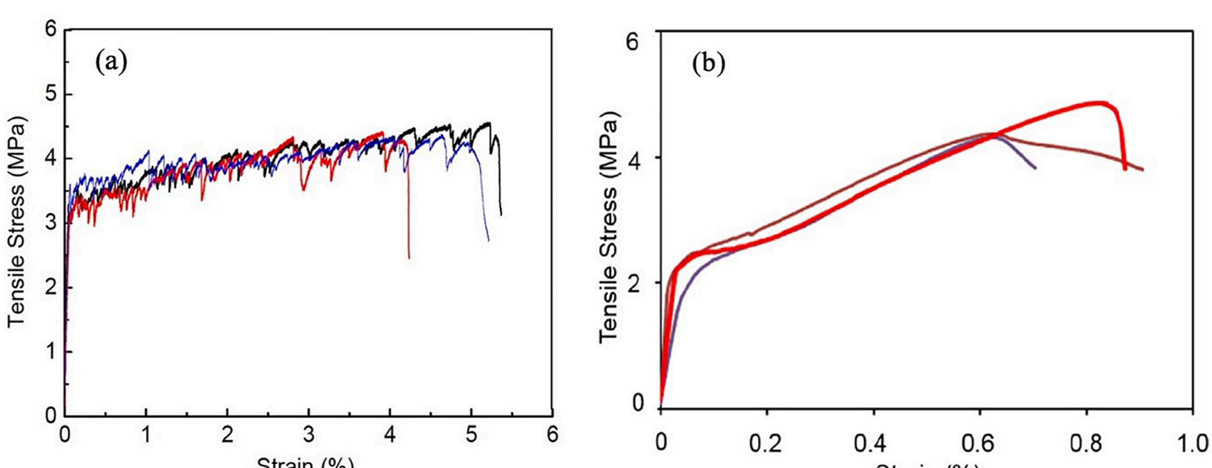

Fig. 11. Comparison on tensile stress–strain curves of (a) PVA-ECC and (b) BF-ECC [165].

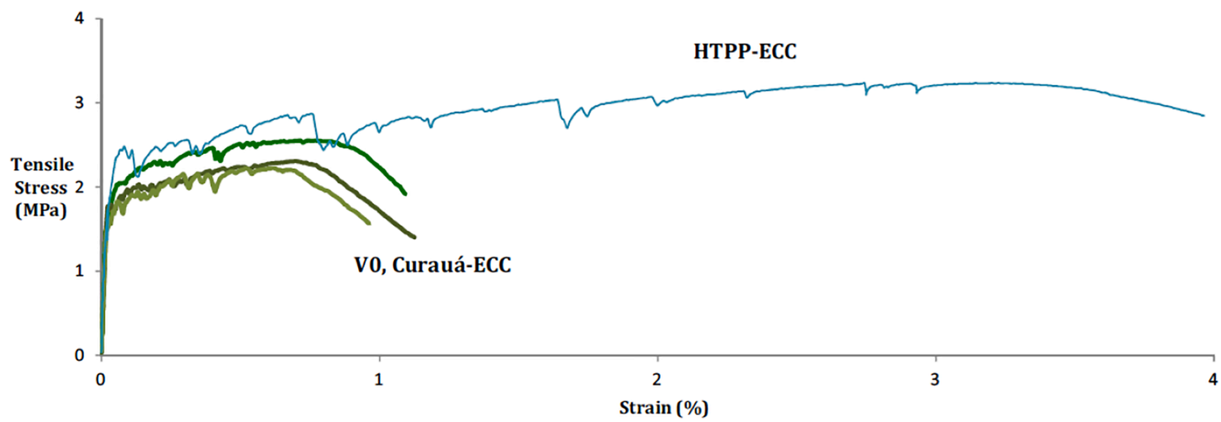

Fig. 12. Tensile stress vs. tensile strain for a Curauá-ECC with three samples compared with a typical performance of high-tenacity polypropylene (HTPP)-ECC [169].

shows a Curauá-ECC with a tensile strain capacity of less than 1%, compared with over 3% for a PP-ECC.

Plant fibers generally have lower density and thermal conductivity [188,190–192]. These properties imply that plant fibers can play a certain role in non-structural elements such as building cladding or facade, where no significant loading is expected. Concerns of low durability [170] of plant fibers in an alkaline environment in the cementitious matrix can be addressed to some degree by either proper fiber treatment or matrix modification [159,188].

4.3. Fiber hybridization

4.3.1. Polyethylene terephthalate (PET) fiber and PVA fiber

Polyethylene terephthalate (PET) is widely used in plastic products, especially in the food and beverage industry [193,194]. A significant amount of PET is disposed of in landfills as waste at end of life (e.g., more than 75% of 32 megatons of PET product in the U.S. ended up in landfill in 2018 [195]), even though the recycling of PET has been gradually increasing.

Yu et al. [171] and Lu et al. [172] attempted to incorporate PET (virgin recycled or oil-coated recycled) as a means of simultaneous greening and cost-lowering of ECC. The PET fibers are spun-drawn from recycled PET melts. The interfacial friction bond is low due to its smoother surface. Surface treatment for PET fiber with NaOH solution and a silane coupling agent delays the crack width development of the composite material, as shown in Fig. 13. That said, the fraction of PVA and use of PET fiber were recommended to be 1.0 vol% PVA fiber and 1.0 vol% PET fiber, regardless of the surface treatment, within several hybridization ratios of 2 vol% in total. The tensile strength and strain capacity are limited to less than 3 MPa and 2%, respectively.

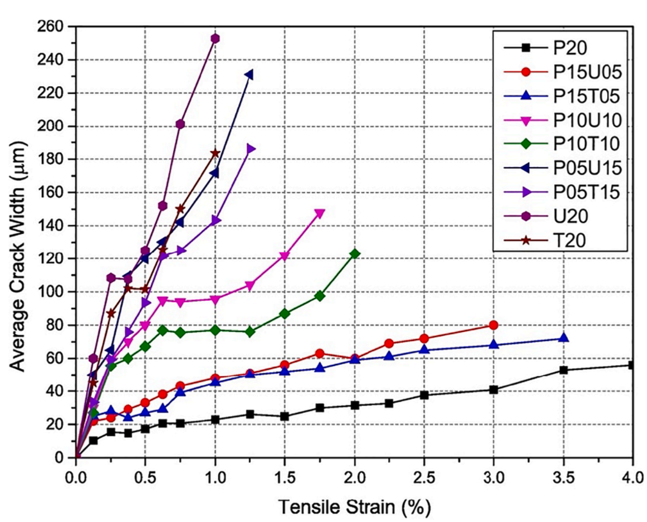

Fig. 13. Crack width development of the hybrid PET-PVA-ECC. P: PVA fiber, U: untreated PET fiber, T: treated PET fiber. Numbers next to each symbol stands for the volume fractions mixed in the ECC mixture (e.g., P10U10 accounts for the case of 1.0 vol% of both PVA and untreated PET fibers). (Adapted from [171]).

The embodied energy and cost of the hybrid PET-PVA-ECC showed over 40% reduction, while CO_2 emission was reduced by more than 50% of those of typical ECC with PVA fiber only. These green credentials could be diminished if additional processing including surface treatments are employed to improve the PET fiber. The significant need to recycle waste PET from consumer products provides impetus to further investigations of PET fiber and ECC containing PET hybrid with other fibers.

4.3.2. Basalt fiber (BF) and PVA fiber

Özkan and Demir [166] studied ECC with hybridized PVA and basalt fiber (BF). The cost of ECC mixture including BF decreased by up to 35% as the BF content increased (i.e., 25, 50, 75% substitution of PVA fiber by weight). However, the mechanical properties of the BF-PVA-ECC were significantly impaired, except for compressive strength [196,197]. The positive effect of BF addition on the compressive strength was attributed to the formation of a load-bearing skeleton of stiff BF. The tensile and flexural properties were negatively affected by the brittle unoiled BF that possessed a high chemical bond and suffered premature breakage at inclined angles [198,199] (Fig. 14).

Despite an increasing amount of literature on hybrid fiber ECC, either for purpose of reducing carbon and energy footprints or for improving composite mechanical properties, the progress has been relatively limited. This is likely a result of a misunderstanding of the mechanics of ECC with a hybrid fiber system. It is recommended that future studies make better use of the σ-δ relation of the individual fiber and the hybrid system. The σ-δ relation embodies the physical (length, diameter) and mechanical (tensile strength, stiffness, bond characteristics) information of the fibers, and is directly linked to the composite strain-hardening properties as well as crack width [1].

5. Summary and conclusions

The urgent need to address climate change by lowering the carbon footprint of the built environment has prompted the continuous development of greener ECC composites over the last decade. ECC has significant potential to reduce the carbon footprint of infrastructure by eliminating repeated maintenance associated with concrete fracture and steel corrosion in reinforced concrete elements. However, the embodied energy and carbon in ECC material can and often exceed those of ordinary concrete. This recognition prompts the increasing number of

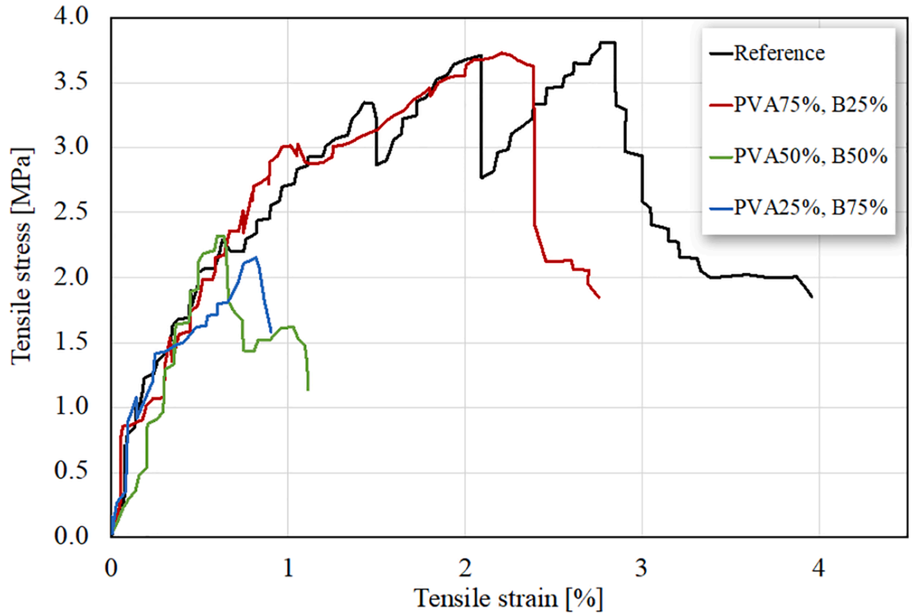

Fig. 14. Tensile stress – tensile strain curve of the BF-PVA-ECC (adapted from [166]). PVA/B#% stands for the mass fraction of either PVA fiber or basalt fiber, amounting to the total mass of 26 kg per unit volume of the mixture (m^3). Reference is PVA-ECC (i.e., PVA100%, B0%).

investigations into alternatives to OPC, manufactured sand, and PVA fiber while maintaining the inherent mechanical or durability properties of PVA-ECC, treated as the baseline ECC in this paper. This paper summarized and analyzed the available body of literature on greener ECC development, to define the current state-of-the-art of this subject, and to serve as a guide for a future trajectory for truly green ECC. This literature review has the following findings:

Fly ash (FA) remains the most widely used supplementary cementing material (SCM), serving as a green binder itself, and as an aid to enhancing the performance of other SCMs. The aid provided by FA includes the suppression of alkali-silica reaction (ASR) associated with hollow glass microsphere (HGM) in a cementitious binder, enhancement of mechanical and eco-friendly properties in MgO-included ECC, and maintenance of tensile ductility in the presence of solid waste ceramic powder (CP) in the binder. FA has also been used for the complete replacement of OPC in engineered geopolymer composites (EGC). The concern over the potential shortage of this highly attractive material prompts the exploration of promising SCM alternatives in OPC-based ECC. Recent advancement of limestone calcined clay (LCC)-based ECC as well as glass powder (GP)-CP-based ECC appears promising.

The replacement of manufactured silica sand by natural sand including sea-sand, river-sand, and dune-sand as well as recycled waste aggregates holds promise to promote sustainability through the use of local ingredients and industrial waste streams. Natural sands are characterized by physical parameters (i.e., particle size, sphericity, or roundness). The sand particle size affects strength-related mechanical properties, while the remaining morphological parameters influence the overall mechanical properties. While the replacement of manufactured sand by natural sand can be considered successful, the interfacial chemical interaction between the green sand / recycled waste aggregate and binder ingredients in the composite needs to be further clarified.

The increasing availability of local man-made, natural, and waste-derived fibers offers exciting greener/lower-cost alternatives to the traditional PVA fiber used in ECC. However, the newer fibers must be properly evaluated for mechanical and durability properties of the fiber and the resulting ECC. The multi-dimensional demand of eco-friendly, fresh, and hardened properties, and long-term durability performance of ECC is balanced by the broader availability of fiber types and by the almost unlimited possibilities offered by fiber hybridization. Localized fibers include PVA, PE, and PP fibers, natural fibers include basalt and Curauá plant fibers, and recycled waste fibers include PET fiber. While holding significant promise, progress in ECC with hybrid fiber systems requires better use of micromechanical design basis, moving beyond the current trial and error approach.

The full menu of greener fiber, greener aggregate/sand, and greener binder, and their selective combinations opens a vista of emerging greener ECC with low embodied carbon that dramatically lowers use-phase carbon footprints of civil infrastructure through its ductility and self-controlled crack width. Building on accomplishments over the last decade, future success in this effort will contribute to the promotion of sustainable infrastructure with low embodied and operational carbon. This will bring humankind one step closer to harmonizing the built and the natural environment.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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