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Bulk fillers from food waste for polymeric bio-composites: The influence of filler type, particle size and volume ratio on furan-matrix composites

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A B S T R A C T

Bio-based composites provide promising low embodied-carbon alternatives to technical materials, but they generally rely on virgin biomass which raises concerns about agricultural land use for non-food crops. Bio-composites made from organic waste address these concerns by providing high carbon-sequestration opportunities with fewer virgin resources. But the sourcing of these waste streams and their impact on the mechanical and functional properties of the bio-composite are poorly understood. This study investigates food industry waste as bulk fillers in bio-composites with a furan resin matrix. Six waste streams were selected based on local availability and current underutilisation. Firstly, bio-composite samples for each filler type were prepared and tested for strength, water absorption and freeze-thaw resistance. Secondly, the two most promising fillers, walnut shell and spent coffee ground, were investigated further, assessing the influence of filler particle sizes and filler content fraction on bending and impact strength. Finally, a carbon impact analysis of the primary production and fabrication was performed to evaluate the carbon footprint of the developed bio-composites, compared to conventional construction materials. With a mean bending strength up to 58 MPa the walnut shells and spent coffee fillers produced the highest performance bio-composites, while variants of cacao bean shells and cherry pits showed blisters and cracking, resulting in lower mechanical properties and higher water absorption. Walnut-based composites benefited from a blend of grain sizes by improving packing density, requiring less resin, while maintaining mechanical performance. The carbon impact analysis showed that a bio-composite with 55 % walnut shell filler is a low-carbon alternative to construction materials such as ceramics, aluminium and steel within the considered life-cycle phases and use case. The findings demonstrate the feasibility of utilising food-industry waste in bio-composites and present the further research needed in the development of these more sustainable materials.

1. Introduction

The need for more environmentally friendly building materials raises the new challenge of limited availability of virgin renewable materials due to slow regeneration of the bio-based resources, land use challenges (e.g. loss of biodiversity, displacement of food / feed crops, etc.) [1]. Polymer based composites made from waste products and in particular bio-polymer composites incorporating (organic) bio-waste, offer an attractive alternative for replacing high-carbon impact materials in construction: a renewable material made without direct extraction of virgin grade materials, with carbon sequestration characteristics, and with significant reductions in waste treatment, landfilling and associated greenhouse-gas emissions.

There are diverse possible waste streams that could provide biobased raw materials for bio-composites for different end-applications, including animal byproducts as well as plant parts from the food processing industry [2]. This waste could potentially be used as a bulk fillers, which often constitute a significant portion of composites and therefore play a significant role in the overall environmental impact [3]. The present study therefore focuses on the potential use of organic waste products from the food industry as bulk fillers in a furan-based bio-composite for load bearing applications in weather-exposed conditions. The focus is not on maximising strength-to-weight or stiffness-to-weight ratios (as is often the case for high-performance aerospace applications), but rather to assess the suitability of such composites for semi-structural applications in the built environment e.g. façade panels, urban furniture, signage, roof shingles/gutters or elements for green infrastructure. Such applications require moderate strength and stiffness in addition to

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shapeability, surface quality, environmental durability and cost-effectiveness [4].

Polymer matrix composites (PMCs) are made by combining a polymer matrix with a fibrous reinforcements and/or particulate fillers [5]. The particulate filler phase is typically stronger than the matrix. It provides strength and stiffness, while the matrix binds the fillers and distributes applied loads across the structure [5]. Polymeric composites can be fabricated by injection moulding, compression moulding, extrusion, contact moulding (or hand layup) and rotational moulding [6]. The choices of fabrication process, matrix and fillers, end-product properties and production scale are interdependent.

The principal constituent materials of PMCs are as follows:

- Polymers, which can be either thermosets or thermoplastics. The latter soften with heat and can be (re)moulded into a desired shape, whereas the former are cured at elevated temperatures and become permanently rigid after cooling [5]. Thermosets are formed by condensation polymerization and usually have higher thermal and chemical resistance than thermoplastics [7]. Polymers that are made fully or partially from biomatter and/or are bio-degradable are called bioplastics.
- Fibres, that can be added to polymeric composites for reinforcement as either short or continuous fibres. The latter can be in unidirectional, multidirectional or woven form [6]. Fibres can be either synthetic or natural.
- Granular or powdered fillers, mineral- or bio-based, which are predominantly used to achieve dimensional stability and reduce the cost of end products, in which case they are known as bulk fillers or extenders [8].
- Functional fillers, which are added in small quantities to achieve particular material characteristics such as colour, electrical conductivity, ultraviolet (UV) resistance or improved processing [9].

Bulk fillers used in polymers are typically mineral-based powders such as calcium carbonate, talc (hydrated magnesium silicate) or carbon black (paracrystalline carbon), in particles smaller than or equal to 10 μm [10]. But bulk fillers may also be bio-based. However, since it is difficult to grind organic matter finely, bio-based fillers are typically used in larger particle dimensions than their mineral counterparts [10]. There is no strict boundary between matrix-dominant bio-composites and other bio-composite materials such as particleboard or chipboard, that contain smaller fractions of polymer binder resulting often in high-porosity and light-weight products [4].

Several studies have investigated the use of bio-based fibres in composites, including attempts to substitute cultivated fibres flax and hemp with agricultural residues such as sunflower stalk, corn stalk, and sugarcane bagasse [2,11]. Waste-product based fillers have also been proposed as alternatives to conventional mineral fillers. For example, periwinkle shells have been incorporated into recycled polypropylene (PP) [12] and eggshells have been tested in epoxy composites [13]. A comparison of peach pits and pine wood particles in PP showed that peach pits reduced moisture uptake but also lowered mechanical performance [14]. Similarly, cherry pits were compared with wood particles in PP; in this case, poor mechanical properties were attributed to fatty acid compounds interfering with interfacial bonding [15]. Walnut shells have been studied as fillers in both epoxy resin [16] and polyester [17], where they were found to increase hardness. Overall literature indicates that plant-based fillers can offer specific benefits, while downsides like increased moisture uptake or lower mechanical strength will occur at increased filler volumes.

Advances in bio-polymers have enabled fully bio-based composites, shown in case studies such as the EU-funded BIOBUILD project with a furan resin façade reinforced by flax and jute fibre [18], and the Basajaun project, which uses wood fillers in a bio-based polymer with basalt reinforcement to produce wall modules [19].

This study focuses on bio-composites composed from bio-based fillers from food industry waste streams in the Netherlands and a bio-based thermoset furan resin, fabricated by a compression moulding process. The study was guided by a dual objective: to devise bio-composites with useful mechanical and functional characteristics for applications in the construction sector, and to achieve a low environmental impact through the use of a high amount of bio-waste filler. It endeavours to provide a systematic basis for comparing the different waste-based filler materials, it identifies methods for incorporating them successfully into bio-composites and it evaluates the influence of filler particle size and filler-to-resin ratio on the mechanical properties of the composites. This is achieved by first evaluating several underutilised food industry by-products for their potential as bulk fillers in Section 2, followed by the experimental investigations and desktop carbon impact analysis in Section 3 and the corresponding results and discussion in Section 4 and Section 5, respectively. The overarching conclusions and recommendations for future research are presented in Section 6.

2. Material selection

The functional and mechanical properties of PMCs are highly dependent on the interaction between the fillers and the matrix, which is in turn influenced by the filler characteristics (such as particle size, shape, surface texture, porosity, chemical structure), and the filler-to-matrix ratio [8,20]. The choice of fabrication method, which can have an influence on the surface finishes, dispersion and bonding of the materials, is steered by the characteristics of the constituent materials (matrix and filler) and by the desired shape and form of the end-product.

2.1. Bio-based bulk fillers

The bio-composites in this study focus exclusively on cost-effective granular bulk fillers i.e. it excludes continuous fibres such as those used in high-performance applications for wind turbine blades or aircraft components. The functional and mechanical properties of the bio-composites were evaluated against the requirements for load bearing applications with weather exposure such as façade cladding, roof tiles and similar applications in the built environment, with a focus on strength and resistance to impact, moisture and frost.

The food-processing industry typically generates large amounts of waste materials with low economical value, amounting to 21 million tonnes of pre-consumer waste in the European Union annually (Fig. 1). The various agricultural and industrial leftover can be plant parts, animal based waste as well as non-edible by-products from processing or rejected produce, with fruit, vegetables, nuts and oil crops being the largest waste producing streams [21].

The Netherlands is one of the largest food exporting countries in Europe and produces over 6.5 million tonnes of pre-consumer waste annually from the food, beverages and tobacco industry [28]. Forty percent of food industrial waste is unfit for human or animal consumption and is fermented into fertiliser, used for the production of bio-gas or is thermally recycled (bio-fuel) [21,29]. Unlike thermal recycling, fermentation and landfilling, the absorption of waste streams in bio-based products slows carbon release into the atmosphere [30], therefore engineered building products from waste-based bio-composites are a preferable route of recycling.

Food waste streams in the South Holland region of the Netherlands were reviewed based on the following criteria:

- Processing potential: grindable, non-fibrous solids suitable for granular fillers.
- Sustainability: locally available and underutilised waste streams with short transport distances.
- Up-scaling potential: scalability for future production applications.

The following waste streams were identified:

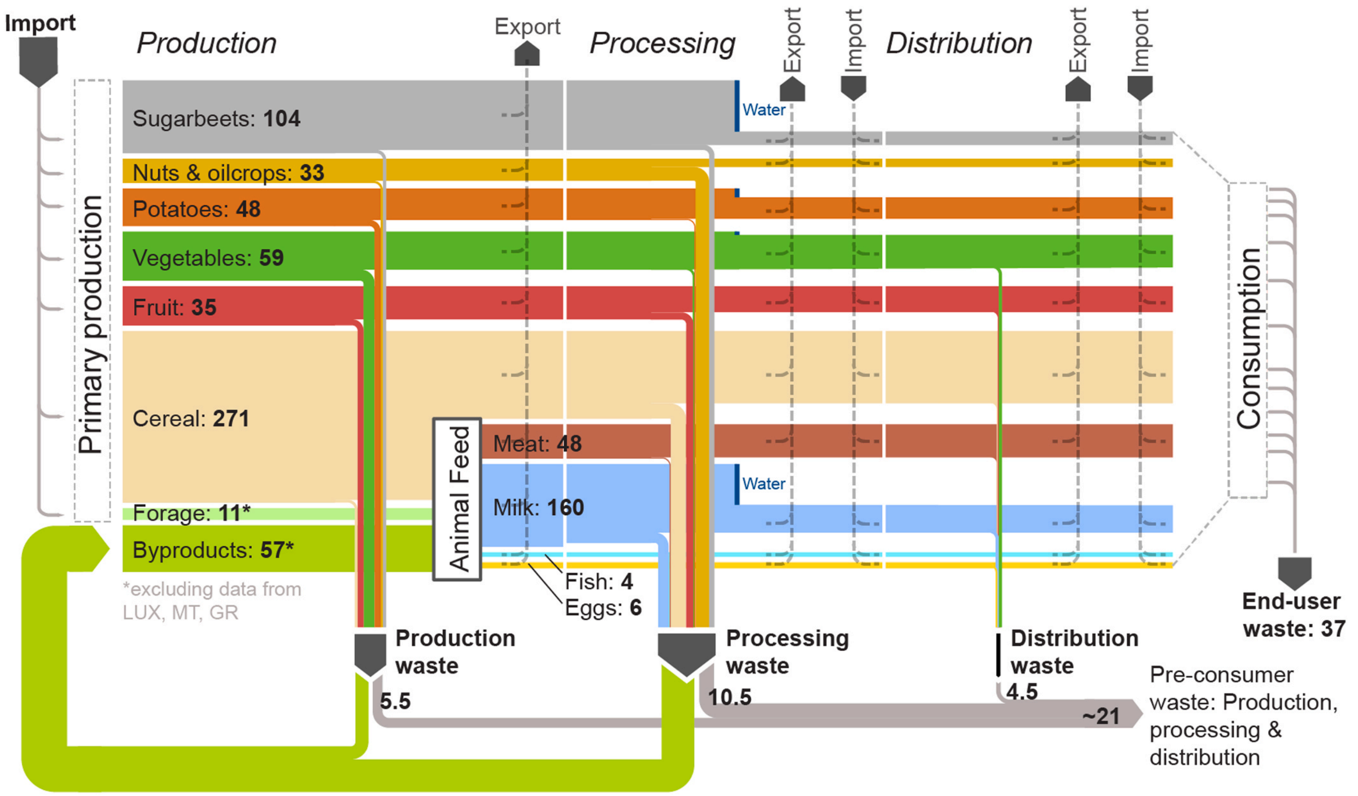

Fig. 1. Food flow diagram and estimated food-waste generated in 2022 in the EU, in Mt, based on [21–27].

- **Coffee grounds**: Coffee grounds are major waste source since coffee is traded and used worldwide at a large scale. With no functional value as animal feed, the grounds are typically thermally recycled or used as soil improvers [29].
- **Cacao bean shells:** Cacao bean shells are a residual product obtained from roasting and crushing cacao beans, that is typically discarded or recycled into low-value applications, including biofuel, fertiliser, and has limited use as feed for animals [31]. The bean shell is a waste product commonly available in the location of chocolate production. It differs from the edible cacao fruit hull, which is typically removed from the beans in the country of cultivation. The shells contain compounds such as dietary fibre, antioxidants and vitamins. Therefore besides the potential use as engineered bio-materials, cacao bean shells are also useful for other value-added products such as cosmetics and pharmaceutical products [32].

- **Cherry pits**: Cherry pits, like other stone fruit pits, which are extracted in the production of canned products and jams, can also be used in cosmetics, energy generation and as grit for pressure washing, [29]. Most stone fruit pits consist of an inner stone and an outer shell. The inner stone can be utilised in the production of cosmetics but often the whole pit is thermally recycled due to its high calorific value [14].
- **Walnut Shells**: Walnuts have a hard chemically inert outer shell, which makes it attractive for recycling. WS can be used as an abrasive for polishing and is effectively used for filtering oil and heavy metals from water [33].

Fig. 2. Raw filler material as received, SCG and DCG dried.

2.1.1. Materials sources

From the identified waste streams, six different fillers were obtained (Fig. 2). All fillers are by-products from the food industry that were locally available in South Holland: The spent coffee grounds (SCG) were collected from a local café in a wet state and subsequently dried in an oven at 90°C for about 4 h. A second variant, de-oiled coffee grounds (DCG) were obtained from a company utilising the coffee waste products for cosmetics (Caffe.inc, Amsterdam, NL) in a dried state. Sold as a by-product of cosmetic coffee-oil, the de-oiled grounds contained, both rejected ground coffee and spent coffee grounds of an unspecified ratio from which then oil was extracted [34]. Cacao bean shells (CS) and roasted cacao bean shells (RCS) were supplied by a cacao processing company (Crown of Holland BV, Middenmeer, NL). The two types of shells, unroasted and roasted, came from two different production lines within the same company, where in one line the shells are extracted before the roasting process. Both the walnut shells (WS) and cherry pits (CP) were obtained though small-scale private orchards within the Netherlands.

2.2. Bio-based matrix

Due to the relatively demanding exposure conditions expected for the possible end applications in the built environment, the thermoset furan resin was chosen as the matrix material in this study. Furan resins are widely used in applications requiring high durability and chemical resistance, such as industrial coatings, sand binders for metal casting, and foundry cores [35]. Their fully renewable origins, combined with their resistance to moisture and thermal degradation, makes them a promising choice for a broad range of bio-composite products.

Furan resin, or furfural alcohol resin, is derived from renewable biomass. It cures at indoor ambient temperature with the addition of a strong acid as a catalyst and the curing speed increases at higher temperatures [36]. In this study, furan resin, supplied by TransFuran Chemicals, was used throughout. The resin has a density of approximately 1.21 g/cm^3 and was cured using a (2-Hydroxyethyl) ammonium nitrate catalyst. The catalyst was mixed with the resin at a mass ratio of 1:20. Linseed oil was added at a ratio of approximately 1:55 to the BMC and was additionally applied to the mould to facilitate demoulding. The selected filler and matrix are listed in Table 1.

2.3. Fabrication

Compression moulding offers a high degree of design freedom and produces high quality surfaces, making it well-suited for the fabrication of products that are visible at close range (e.g. façades panels, street furniture etc.). The fabrication process can accommodate various recipes, filler particle sizes, and is suitable for both particulates and non-woven fibres. The moulding of a thermoset based composite typically starts by creating dough, called bulk moulding compound (BMC), by mixing the resin with the filler ingredients and pre-curing it, followed by fully curing the BMC under pressure (here 6 MPa) in a mould by means of a heated press, to take the shape of the final product (Fig. 3) [2].

2.4. Composition parameters

Two constituent parameters, filler particle size and volume fraction, are of interest in the present study. They play a critical role in the properties of a composite end-product and are often considered in composite research [10,37,38]. Their influence varies with the specific materials and fabrication methods used, but the following common trends and causal relationships are identified:

- **Filler particle size (grain size):** The size of filler particles significantly influences the mechanical and functional properties of composites by effecting bonding with the polymer matrix, load transmission, and additive dispersion [38]. For bio-based fillers, particularly hydrophilic or porous types, grain size plays a crucial role in water absorption. Larger filler sizes often lead to higher water absorption rates, as surface connected particles absorb water and larger particles can introduce more moisture into the composite [14,16]. Smaller fillers generally enhance stiffness and Young's modulus by improving packing density and interfacial bonding, whereas larger fillers can lead to agglomeration and void formation, thereby reducing mechanical performance [20,38]. Fabrication methods further modulate the influence of filler size. Hand lay-up methods produce larger voids (air inclusions) with larger grain sizes, but this is less pronounced in compression moulding, where the applied pressure helps to expel such air inclusions and leads to a denser structure [16].
- **Particle sizes distribution:** In production of other filler-based composite materials such as concrete, mixing fillers (aggregates) of different sizes optimises packing density, minimises surface area and reduces binder (cement) usage while maintaining a suitable viscosity (known as workability in concrete technology) [39]. In most PMCs, packing density depends on the size distribution of filler particles from the milling process, with no additional adjustments. However, systematic mixing of filler sizes in PMCs can reduce resin usage by 20–30 vol% [37], though this method is mainly applied in high-performance composites such as dental materials. In this study, size distribution effects are assessed by adapting the Fuller and Thompson formula (1907) [39], which is commonly used for concrete mix design.
- **Filler load:** PMCs typically contain 20–90 % bulk filler, depending on the matrix and intended function. Increasing filler content generally increases the stiffness and enhances the dimensional stability of the end product [40]. In thermoset compression moulding, a minimum filler load is required to stabilise the BMC and prevent resin spillage. Filler particles larger than 50 μm can be incorporated up to 64 % while maintaining matrix continuity [37]. When the fillers deform under pressure, matrix continuity can be maintained at a maximum of 94 % filler content [37]. The influence of an increasing filler content on mechanical performance depends on the filler size, shape, and hardness relative to the matrix. Higher filler ratios reduce costs and shrinkage during production [8], while waste-based fillers offer the additional benefit of reducing the reliance on virgin materials, thereby reducing the embodied carbon of the composite.

Table 1
Selected waste types and matrix material.

Function	Material	Abbreviation
Waste-based filler	▪ Spent coffee grounds	SCG
	▪ De-oiled coffee grounds	DCG
	▪ Cacao bean shells	CS
	▪ Roasted cacao bean shells	RCS
	▪ Walnut shells	WS
	▪ Cherry pits	CP
Matrix	▪ Furan resin	

3. Method

3.1. Filler characterisation

To better understand the properties of the grinded filler materials, the following key characteristics influencing bio-composite formation and processability, were analysed.

Solubility tests were performed to asses water absorption and leaching behaviour of the fillers. Although solubility in water does not directly translate to solubility in furan resin, it highlights differences between waste streams that may affect filler-matrix interactions or surface quality. Density measurements provided an indication of porosity, which influences workability and resin spillage during moulding. The milling behaviour was evaluated to judge processing feasibility, while the particle morphology was examined for insights on possible packing density, shape related stress concentrations and their possible correlations with mechanical performance.

- **Water solubility**: The mass of soluble compounds of the different filler materials was determined by hot filtration. The fillers were powdered ($\leq$250 μm) and mixed with water at 95°C to 100°C, followed by filtering through 10μm filter paper (Hario V60 bleached filter paper). The filtrate was re-filtered repeatedly until no discoloration of the filtrate was visible. The residual material was dried at 90°C for 5 h and weighed.
- **Density**: The density was determined by the water displacement method. Approximately 10 g of filler was added to a graduated beaker containing 10 ml of water and the increase in volume was recorded. The reading precision was ±0.2 ml, which corresponds to an uncertainty of ~2–3 % across the measured density range. Possible absorption of water by the filler was not corrected for, so the

Fig. 3. (a) Milled and sieved walnut shell filler; (b) open kneader; (c) bulk moulding compound pre-pressing; (d) hydraulic press.

reported values should be regarded as indicative rather than absolute.

- **Ease of milling and particle shape**: Visual observation of agglomeration during grinding and sieving operations and optical microscopy of representative filler samples.

3.2. Sample production and variants tested

Bio-composite specimens were produced as follows:

1. Milling of filler material with a ball mill (Fritsch, Pulverisette 6 classic line).
2. Sieving the milled material in a sieving tower (Fritsch, Analysette 3 pro) into grain sizes ≤ 125 *μ*m, > 125–250 *μ*m and > 250–500 *μ*m. The grain size distribution within each sieving step was not assessed further.
3. Drying the filler material in an oven at 110°C for 2 h.
4. Mixing of ingredients and pre-curing of the resin in a kneader with two rotating arms (Linden, Type K Double-Z-Kneader, 1.2 l capacity) heated to 95°C, by adding first the resin, then the catalyst and the linseed oil and lastly the filler. The ingredients were added one at a time, in intervals of approximately 10 min, to allow for the evaporation of access moisture before adding the filler powder, which was supported by the kneaders agitation (≈30 rpm) and air extraction.
5. Pressing the resulting BMC at 6 MPa and 150°C for 15 min in a hydraulic press (Bucher model KHB, 100 ton), thereby producing flat sheets of 5 mm thickness and subsequently cooling down at ambient temperature (21 ± 2°C) and humidity (55 ± 5 %).
6. Cutting the sheets by computer numerical control (CNC) milling into test specimens measuring 80 mm by 10 mm, 80 mm by 15 mm and 50 mm by 50 mm for the Charpy impact testing (HY Tech, Charpy impact tester HY4251), the three-point bending with a universal testing machine (Instron, 5969 testing system) and the water absorption/ freeze-thaw testing, respectively.

Sixteen bio-composite variants were created in iterative steps by one-factor-at-a-time combinations of the three independent variables (i.e. filler type, particle size, and filler content) as shown in Table 2. The recipe details including filler content are shown in Table 3. In the first

Table 3
Bio-composite recipes.

Component	Description	Composition [wt%]		
		Recipe 1 (45 % filler)	Recipe 2 (35 % filler)	Recipe 3 (55 % filler)
Resin	Furan resin	50.76	60.29	41.24
Filler	Waste filler	45.00	35.00	55.00
Catalyst	(2-Hydroxyethyl) ammonium nitrate	2.54	3.01	2.06
Releasing agent	Linseed oil	1.70	1.70	1.70

Table 2
Overview of bio-composite variants and tests performed (three-point bending strength (3P-B); Charpy impact resistance (impact); water absorption (water); freeze-thaw resistance (frost); CO_2-eq emissions analysis (CO_2-analysis).

Tranche	Variant no.	Bio-composite variant characteristics					Tests performed		
		Variant name	Filler type	Particle size [*μ*m]	Filler content [wt%]	Recipe no.	3P-B & impact	Water & frost	CO_2-analysis
1	1	SCG_125_45	SCG	≤ 125	45	1	✓	✓	
	2	DCG_125_45	DCG	≤ 125	45	1	✓	✓	
	3	CS_125_45	CS	≤ 125	45	1	✓	✓	
	4	RCS_125_45	RCS	≤ 125	45	1	✓	✓	
	5	WS_125_45	WS	≤ 125	45	1	✓	✓	
	6	CP_125_45	CP	≤ 125	45	1	✓	✓	
2	7	WS_250_45	WS	> 125–250	45	1	✓		
	8	WS_500_45	WS	> 250–500	45	1	✓		
	9	WS_mix_45	WS	blend	45	1	✓		
	10	SCG_250_45	SCG	> 125–250	45	1	✓		
	11	SCG_500_45	SCG	> 250–500	45	1	✓		
	12	SCG_mix_45	SCG	blend	45	1	✓		
3	13	WS_mix_35	WS	blend	35	2	✓		
	14	WS_mix_55	WS	blend	55	3	✓		✓
	15	SCG_125_35	SCG	≤ 125	35	2	✓		
	16	SCG_125_55	SCG	≤ 125	55	3	✓		

tranche, variants #1 to #6 were first produced and tested for initial comparison of the six filler types (see results in Section 4.1). Here a 45 wt% of filler, with particle size smaller or equal to 125 μm was used. From this first tranche, two filler types, walnut shells (WS) and spent coffee grounds (SCG) were selected for further detailed experiments in a second tranche of testing, wherein bio-composite variants #7 to #12 were used to assess variations in filler particle size for WS and SCG, using the particle size ranges ≤ 125μm, 125–250μm, 250–500μm, and a blend of all three (see results in Section 4.2). Based on the results obtained from tranche 1 and 2 (variants #1 to #12), the most promising particle size for each filler was chosen for tranche 3 (variants #13 to #16), namely: ≤ 125 μm for SCG and "blend" for WS. The rationale for this is further elaborated in Section 4.2. For bio-composite variants #13 to #16, filler contents of 35 wt% and 55 wt% were investigated, and compared to the corresponding 45 wt% variants previously tested (see results in Section 4.2).

The proportions of the three particle size groups used in the blended variants #9, #12, #13, and #14, were determined from Fuller's maximum density criteria [39], as follows:

$$A = 100\ (d/D)^n \tag{1}$$

where A is the percentage passing through the sieve, d is the grain size, D is the largest grain diameter and n is the shape factor ($n = 0.5$ for perfect spheres, $n = 0.4$ for rounded particles, and $n = 0.3$ for angular particles).

Different shape factors were assigned to the filler types based on their particle shapes: $n = 0.35$ for WS and $n = 0.4$ for SCG. This resulted in two slightly different particle size mixtures for each filler, as detailed in Table 4.

3.3. Mechanical and functional testing

The salient mechanical properties and functional performance for the intended applications in the built environment are strength and durability. In particular, this study characterised the bending strength, impact strength, water absorption and degradation after freeze-thaw cycling (Table 5). All specimens were subjected to bending and impact testing. Bio-composite variants #1-#6 were additionally assessed for water absorption and resistance to freeze-thaw cycles. Observations on ease of processing and the visual appearance of bio-composites across the filler types were also recorded in this study. Other characteristics such as UV degradation, resistance to other atmospheric pollutants, fatigue performance etc. were not considered in this study.

3.4. Carbon impact analysis

To provide context for the environmental assessment, the bio-composite variant #14 (WS_mix_55) was selected for comparison with commonly used construction materials for applications like thin façade cladding, namely steel, aluminium, ceramic tiles, granite and softwood. The unit of comparison was global warming potential (GWP) on a per-kg basis and per unit of a simple flat panel product for the life-cycle stages of primary production (A1) and manufacturing (A3) according to EN 15804 [43] (Table 5). The comparison is limited to these stages and metrics as long-term durability and end-of-life properties of the created composites could not be determined at this point. The GWP of reference materials was determined with Ecoaudit tool of the software Granta Selector 2024 R2 [44]. The CO_2-equivalent emissions of the primary production of all constituent materials and the processing emissions for the corresponding sheet products were obtained from literature. The purpose is to illustrate the relative impact of the bio-composite for façade-type applications, and not to evaluate these materials for primary structural use.

3.4.1. A1 – Primary production

The carbon-equivalent emissions of the production of furan resin (assuming an all-fossil energy generation for the production), linseed oil and the catalyst. The filler (WS) was assessed without attributing any carbon impact. In particular: the emissions resulting from the waste-based filler prior to the point of pickup were considered part of the original product's lifecycle (shelled walnuts); additionally, the beneficial CO_2 sequestration of the filler material and bio-based furan resin was excluded from the analysis due to the uncertainties about the life-span of the bio-composite.

3.4.2. A3 – Processing

The CO_2 impact of the production of the bio-composite was assessed by estimating the electrical energy consumption from all processing steps: milling, drying, kneading and moulding. The carbon equivalent of electrical energy was taken as 0.344 kg CO_2-eq per kWh, which corresponds to the average carbon emissions for electrical energy in the Netherlands in 2023 [45]. The assessment assumes a small-scale production, which is likely to differ from an efficient upscaled processing line with a continuous throughput. The energy consumption of the processing machines may also vary, based on the hardness and moisture of the filler supplied and the batch size produced.

In addition to the carbon impact of the bulk materials (kg CO_2-eq/kg), the materials are compared in a hypothetical use-case consisting of a 600 mm by 400 mm cladding plate supported at four points 50 mm inboard from its corners (i.e. support span of 500 mm by 400 mm) with an out-of-plane (lateral) load of 1.3 kNm^{-2} and a maximum allowable out-of-plane deflection of 10 mm. The plate thickness required to satisfy strength and deflection limits of this use-case was calculated for each material; from which the corresponding mass and per-kg carbon impact was computed. Table 6 shows the material characteristics of the bio-composite and the reference materials. Average (i.e. arithmetic mean) values, rather than design values, are used throughout.

4. Results

4.1. Filler characterisation

- **Solubility:** The loss in dry weight (Table 7) shows that the fillers with the largest loss were both variants of cacao bean shells and the de-oiled coffee.
- **Density:** The results of the water displacement are test shown in Table 7 and are indicative of the density. Several fillers (primarily DCG, CS and RCS) are likely to contain a significant fraction of soluble compounds and water absorption was not accounted for. These values should therefore be treated as approximate.
- **Ease of milling and particle shape**: The powdered fillers exhibited distinct shapes and agglomeration behaviours. WS, CS, and RCS were easy to grind and sieve, while SCG, DCG, and CP agglomerated, creating processing challenges. Optical microscopy revealed that the particles of the coffee-based fillers (SCG and DCG) were relatively uniform particles, whereas the ground walnut shells (WS) featured a combination of sharp, semi-fibrous particles and rounded, pebble-like shapes (Fig. 4). The cherry pits (CP) filler consisted of predominantly rounded particles, while the cacao bean shells (CS and RCS) fillers formed thin, flaky particles.

Table 4
Particle size composition for the blended samples (variants #9,#12,#13 and #14).

Particle size bracket [μm]	WS ($n = 0.35$) [wt%]	SCG ($n = 0.4$) [wt%]
≤ 125	61.5	57.5
> 125 – 250	17.0	18.5
> 250 – 500	21.5	24.0

Table 5
Overview of testing methods.

Test	No. of nominally identical specimens per series	Specimen size [mm]	Method/standard
Three-point bending	6	80x15	ISO 14125:1998 [41]
Charpy impact	10	80x10	ISO 179 1:2023 [42]
Water absorption	3	50 × 50	The specimens are submerged in water for 28 days. Weight measurements are taken at 24-hour intervals.
Freeze-thaw cycling	3 (saturated specimens from water absorption testing)	50x50	The specimens undergo 10 cycles of freezing (−17°C for 8 h) and thawing (ambient temperature, 20 ± 2°C for 4 h), after which the samples are inspected for defects and surface changes under a digital microscope (Keyence VHX−7000), and compared to control samples that have not undergone freeze-thaw.
CO_2-eq analysis	-	600 × 400	The global warming potential (GWP) of bio-composite variant #14 (WS_mix_55) is calculated for life-cycle stages A1 and A3 according to EN 15804 [43] and compared with reference materials for flat panel façade cladding.

Table 6
Mechanical properties and calculated thickness and weight of case study panels of different materials.

Plate material	Mean flexural strength [MPa]	Mean flexural modulus [GPa]	Density [kg/m³]	Source	Thickness [mm]	Mass [kg]
WS_mix_55	49.0	5.1	1370	Present study	6.7	1.84
Aluminium	165	70.5	2710	[44]	2.8	1.51
Coated steel	314.0	207.0	7850	[44]	2.0	3.14
Ceramic tile	4.2	5.66	2220	[44]	6.8	3.47
Granite	13.6	59.2	2900	[44]	3.7	3.73
Softwood (fir)	72.6	10.8	443	[44]	13.5	1.30

Table 7
Weight loss of filler materials after filtration and measured density per filler type.

Filler	Weight loss after filtration [wt%]	Average measured density [kg/m³]
SCG	9.0	1089
DCG	23.9	1117
CS	24.4	1070
RCS	27.9	1083
WS	6.8	1470
CP	5.3	1145

4.2. Filler types

The original six bio-composite in tranche 1 (variants #1 to #6), each corresponding to a different filler type were fabricated and tested to assess the effect of different fillers on the bio-composites.

4.2.1. Filler processability

There was a considerable variation in ease of processing across the different fillers. Cacao bean shells (both CS and RCS) were the easiest to mill and sieve, requiring the minimum time for both steps across all bio-composite variants. Walnut shells required approximately 50 % more milling time than the CS due to higher hardness, but no further processing difficulties were encountered. The coffee grounds and cherry pits presented significant challenges. Milling time of SCG and DCG were similar to CS and RCS. The milling of CP took approximately 20 % more time than the CS filler. However, the sieving of SCG, DCG and CP was particularly challenging due to agglomeration on the sieves. The coffee grounds powder exhibited electrostatic attraction, which was overcome by wet sieving the material and drying it after milling. Cherry pits were the most problematic to process; their hard shell and softer core formed a paste-like consistency when ground, that clogged the sieves.

4.2.2. Visual appearance

There were significant variations in the surface quality of the six

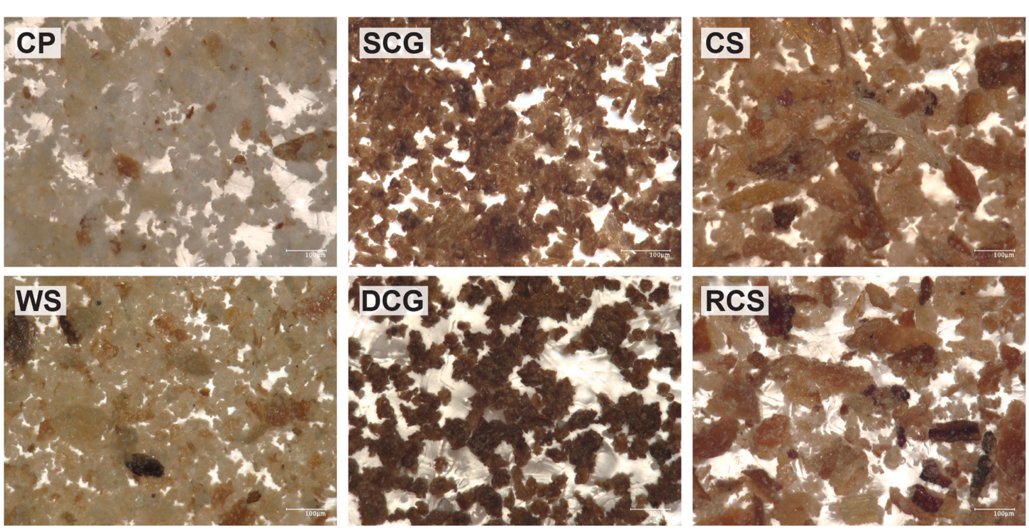

Fig. 4. Filler powder ≤ 125 μm, 1:400 magnification.

variants. The bio-composites containing WS, SCG, and CP fillers resulted in smooth, defect-free surfaces. In contrast, DCG, CS, and RCS variants showed notable surface unevenness and blisters that partially resulted in cracks (Fig. 5).

4.2.3. Bending and impact characteristics

Fig. 6 shows the results of the 3-point bending tests of bio-composite variant #1 to #6. Mean flexural strength ranged from 13.9 MPa (DCG) to 58.7 MPa (WS), and mean flexural moduli varied between 2.8 GPa (CS) and 5.25 GPa (WS) (Table 8). The mean impact strengths of the bio-composites ranged between 1.04 kJ/m^2 (DCG) and 3.3 kJ/m^2 (WS), and followed had a similar rank order to the bending strengths, except for the CS composite which showed a slightly higher impact resistance than the RCS composite (Fig. 6), while exhibiting a comparable flexural strength. Fig. 7

4.2.4. Water absorption

The water uptake by the composites varied significantly across the six filler variants. CP and WS composites showed the lowest absorption rates of 0.8 % in 24 h whereas the highest absorption of 5.5 % in 24 h was recorded in the RCS bio-composite (Table 9). Fig. 8 shows the absorption rates over 28 days. Bio-composites with RCS, CS and DCG filler reached peak saturation during the 28 days and followed by weight loss through leaching, of coloured substances in the water bath.

4.2.5. Freeze-thaw resistance

None of the six bio-composite variants exhibited visible damage from the freeze-thaw cycles. In particular: no loose material (e.g. in form of chips) was formed; no new cracks were developed, and no noticeable warping was observed in the specimens after testing. Discolouration (fading) of the surface and slight loss of gloss were notable after the water absorption test for the CS, RCS, CP and DCG filler types. Microscopic examination of the bio-composite specimens revealed that filler particles became more visible, and the surface had a higher surface roughness after freeze-thaw cycling, compared to unexposed samples (Fig. 9).

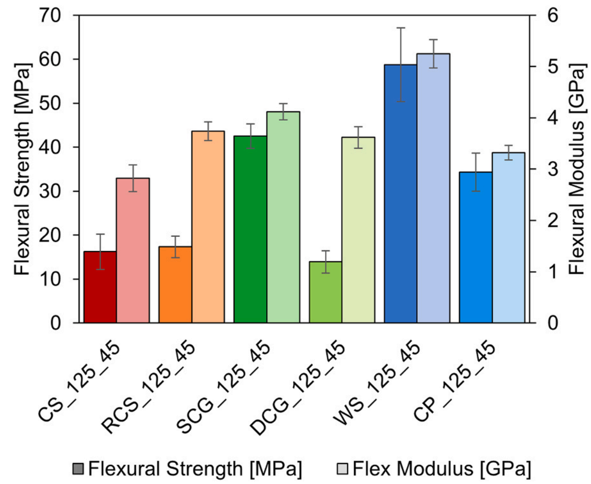

Fig. 6. Flexural strength and modulus of bio-composites variants of different filler types. Whiskers indicate the standard deviation.

4.3. Grain size and composition

4.3.1. Grain size

Bio-composite variants #7 to #12 consist of variations of WS and SCG filler types, in particular, four variations in grain size composition for each of the two fillers (Table 2). During specimen production, it was observed that larger grain sizes occasionally resulted in uneven filler dispersion, with visible filler grains and localised areas with higher resin concentrations, as shown in Fig. 10 (a). Additionally, resin spillage at the mould edges was more pronounced in variants with larger filler particles. However, both issues were not observed in the blended grain size variants, with little to no resin spillage during pressing and an overall homogeneous specimen appearance.

Increasing the grain size for both filler types results in a reduction in flexural strength (Fig. 11). The WS blended particle sized filler resulted in a relatively high flexural strength of 50.6 MPa within the WS filler

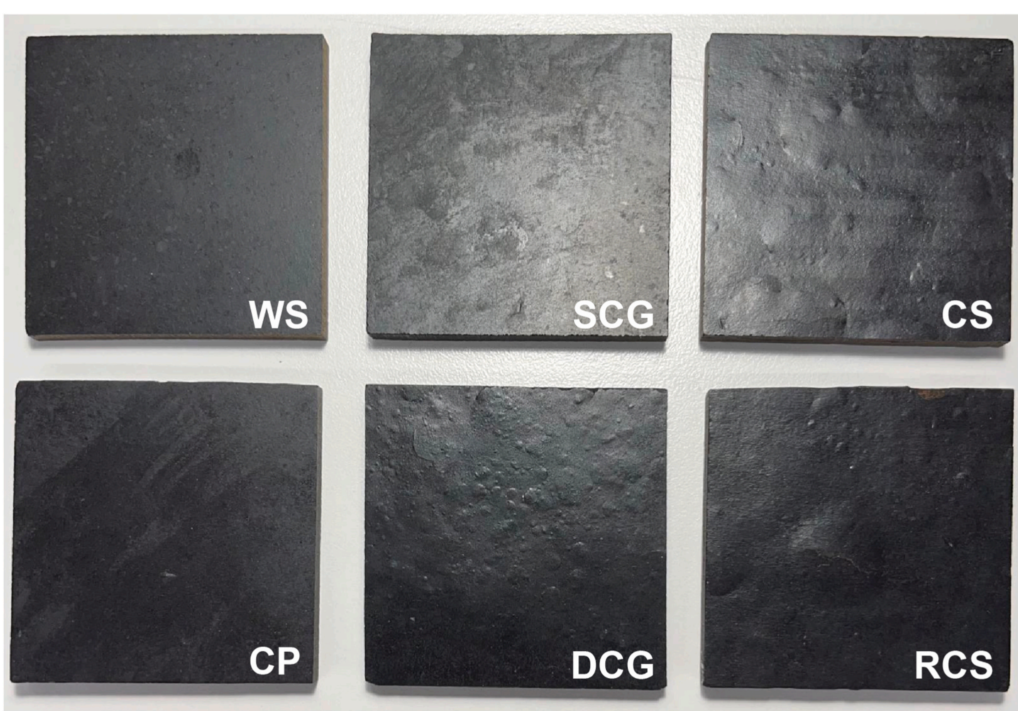

Fig. 5. Samples of bio-composite variant #1-#6 (50 ×50 mm).

Table 8
Test results for 3-point bending test, and Charpy impact test of bio-composite variants #1 to #16.

Variant no.	Variant name	Charpy impact resistance		Flexural strength		Flexural modulus	
		Mean [kJ/m²]	Std dev [kJ/m²]	Mean [MPa]	Std dev [MPa]	Mean [GPa]	Std dev [GPa]
1	SCG_125_45	2.35	0.47	42.52	2.73	4.12	0.16
2	DCG_125_45	1.04	0.40	13.89	2.50	3.62	0.21
3	CS_125_45	1.37	0.46	16.20	3.99	2.82	0.26
4	RCS_125_45	1.06	0.29	17.31	2.45	3.74	0.18
5	WS_125_45	3.34	0.52	58.71	8.37	5.25	0.28
6	CP_125_45	2.42	0.37	34.34	4.32	3.32	0.14
7	WS_250_45	2.61	0.32	49.96	3.88	5.67	0.36
8	WS_500_45	2.07	0.33	41.70	2.02	5.74	0.29
9	WS_mix_45	2.26	0.45	50.61	2.74	5.23	0.20
10	SCG_250_45	1.68	0.27	33.23	3.11	3.48	0.13
11	SCG_500_45	1.79	0.46	31.00	3.81	3.64	0.14
12	SCG_mix_45	1.41	0.12	27.93	3.42	3.42	0.13
13	WS_mix_35	1.63	0.18	35.01	3.06	3.58	0.13
14	WS_mix_55	2.33	0.31	48.98	2.49	5.12	0.24
15	SCG_125_35	2.18	0.45	32.63	3.61	2.98	0.12
16	SCG_125_55	1.53	0.32	32.00	6.20	3.97	0.27

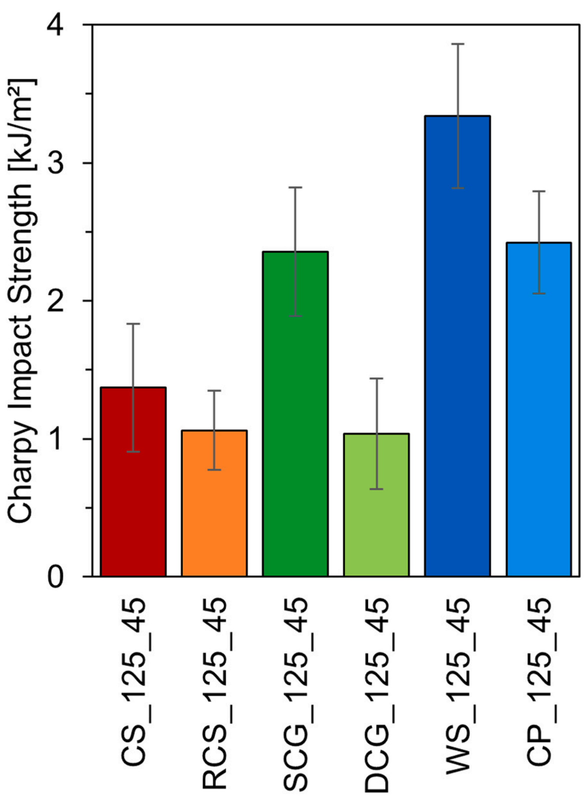

Fig. 7. Impact strength of bio-composites variants of different filler types [in kJ/m²].

Table 9
Water absorption after 24 h and measured maximum.

Filler Type	24 h [wt%]	Max. in 28 days [wt%]
SCG	2.1	9.3
DCG	3.0	13.3
CS	4.3	11.8
RCS	5.5	12.7
WS	0.8	4.8
CP	0.8	4.4

group, but the SCG filler with mixed-size particles showed the lowest flexural strength within that filler type. The smallest grain sizes showed the highest impact strengths for both filler types, while the lowest values

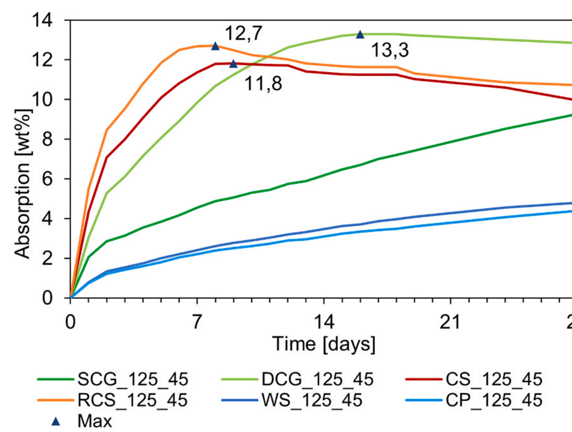

Fig. 8. Water uptake of furan-based bio-composites with 45 % filler content within 28 days.

occurred for the largest WS grain size and the mixed SCG variant (Fig. 12). The scatter in bending strength for the walnut composites was higher for the smallest grain size and lower for larger and mixed sizes, unlike the SCG filler, which showed little variation in scatter across grain sizes.

Only one grain size variant of each filler type was taken forward for the filler content investigation in tranche 3, specifically: SCG_125_45, which exhibited the highest bending strength and impact resistance in its group and was therefore selected for this purpose; and WS_mix_45 which had the second highest bending strength and the lowest dispersion in its group.

4.3.2. Filler content

Bio-composites in tranche 3 (variants #13 to #16) consist of variations of WS_mix and SCG_125 filler types, with ± 10 wt% variations of filler content from the preceding 45 wt%. During specimen fabrication of this tranche, the texture and workability of the BMC varied between bio-composite variants. For the mixed-size WS variants, the BMC consistency remained relatively stable across the filler content range of 35–55 %. In contrast, the ≤ 125 μm SCG variants exhibited notable changes in texture, from a plastic, viscous consistency at 35 % filler to a granular, sand-like texture at 55 % filler.

The results from the bending and impact testing in Fig. 13, show that the 45 % filler samples of both filler types (WS and SCG) have a higher strength than the 35 % and 55 % variants. However, the composite variant with 55 % walnut filler (with mixed grain sizes) performed only

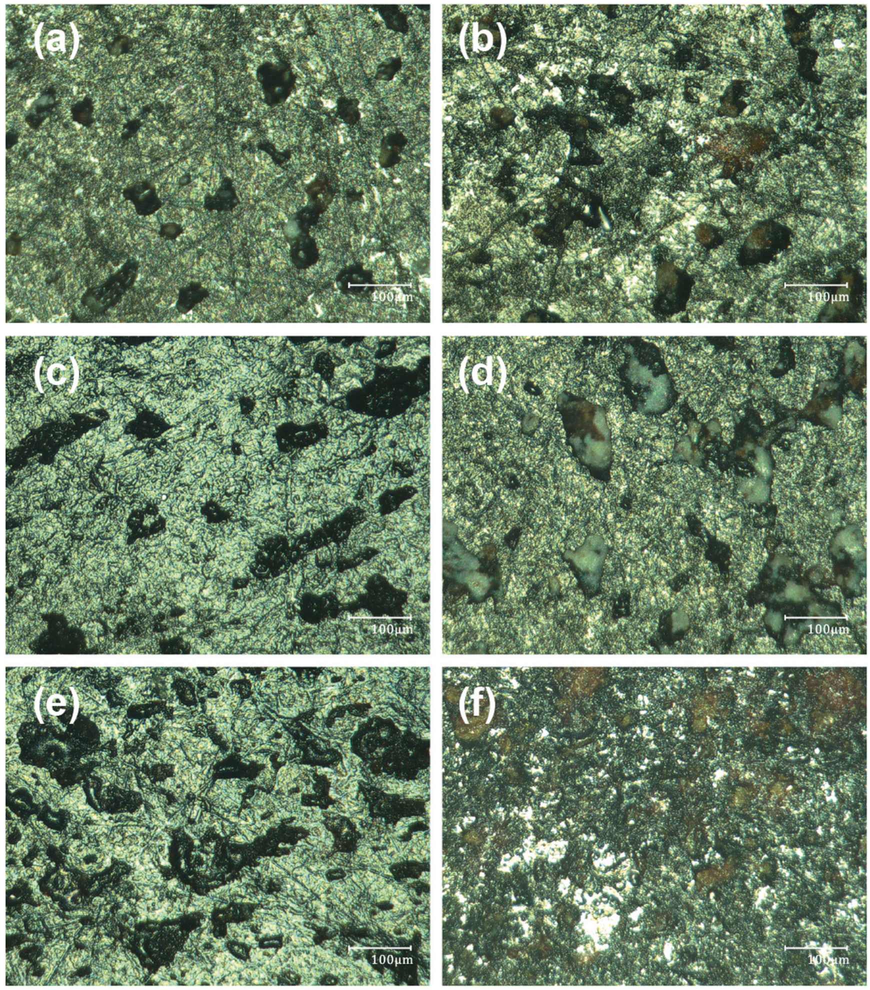

Fig. 9. Surfaces of bio-composite samples before (left) and after (right) exposure to water and frost - (a,b) WS_125_45; (c,d) DCG_125_WS; (e,f) CP_125_45.

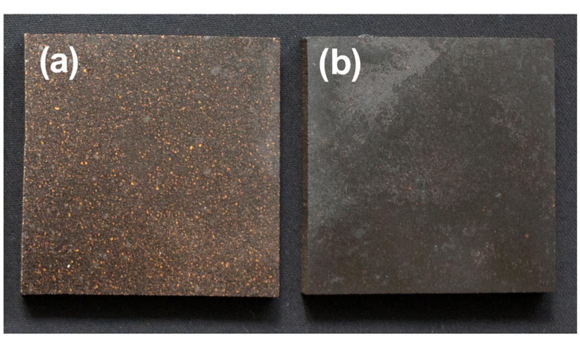

Fig. 10. Difference in appearance of different grain sizes. (a) WS_500_45; (b) WS_mix_45 50x50mm specimen.

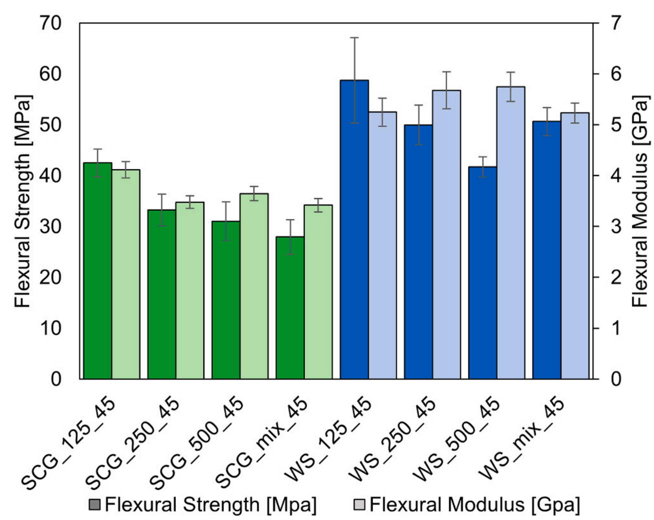

Fig. 11. Bending strength, flexural modulus of specimen with different filler particle sizes.

marginally worse than the corresponding 45 % variant, while the impact resistance increased with filler load. The SCG filler composites with 45 % filler showed the highest performance within that filler group, with a drop in mechanical performance (bending strength, flexural modulus and impact strength) for the 35 % and 55 % variants.

4.4. Carbon footprint

4.4.1. Primary production

The carbon footprint of the primary production of the constituent materials, was calculated by accounting for the impacts of all ingredients, with values taken from literature (Table 10). The carbon-equivalent for the primary production of the bio-composites with 55 % filler is 0.95 kg/kg. For comparison, the resource-related emissions of the WS_mix_55 composite are nine times lower than construction-grade aluminium (9.11 kg CO_2-eq/kg), but more than three times

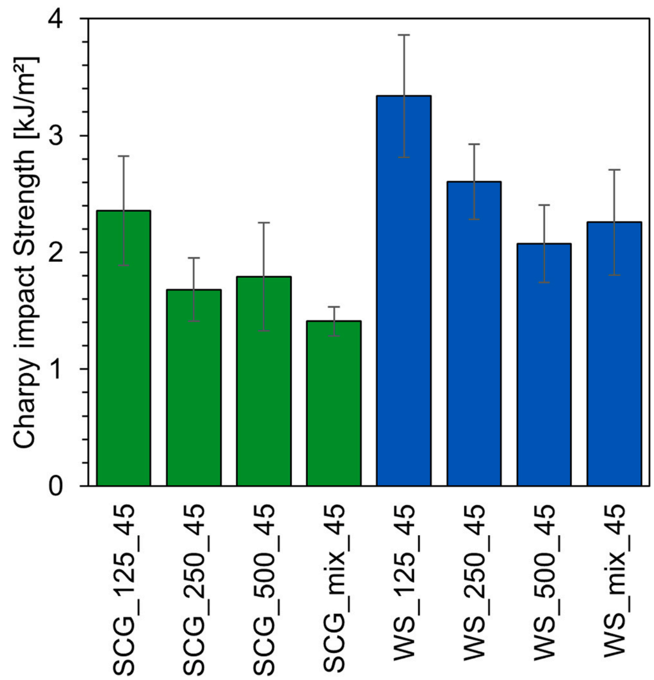

Fig. 12. Charpy impact resistance of samples with different filler particle sizes.

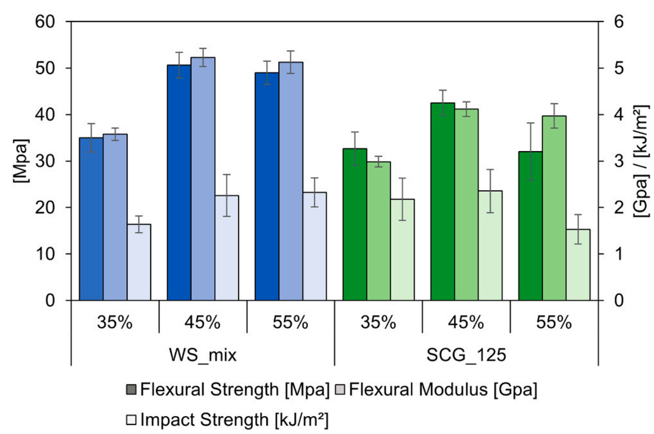

Fig. 13. Bending strength, flexural modulus and impact strength of specimens with different filler content.

higher than softwood (0.27 kg CO_2-eq/kg) [44].

4.4.2. *Processing*

The processing of the bio-composite was calculated for a plate thickness of 6.7 mm, and corresponding plate mass of 1.84 kg, required to support the imposed load within the material strength and prescribed deflection limits (see Table 6), resulting in 0.78 kg of CO_2 equivalent emitted per panel for the processing stage.

Quantifying processing energy is challenging, because it depends on various factors, including the production schedule, processing speed, standby time, and ambient temperatures. The values estimated for each process step are shown in Table 11. On this basis, the heat-intensive step of compression moulding was the largest contributor to energy consumption. Table 12 shows the emissions for primary production and processing per kg of the bio-composite WS_mix_55 compared with the average expected emissions for other typical construction materials for these life-cycle stages.

A comparison with other construction materials on a per-panel basis, shows that the composite WS_mix_55 has a relatively low embodied carbon within the life-cycle phases assessed (Fig. 14). Among the materials considered for this use case, softwood has a lower carbon impact, both in primary production and processing.

Table 10
Carbon impact per recipe.

	Ingredient content [wt%]				Carbon impact bio-composite [kg CO_2-eq/kg]
	Filler	Furan resin	Catalyst	Linseed oil	
Recipe 1	45.0	50.8	2.5	1.7	1.16
Recipe 2	35.0	60.3	3.0	1.7	1.37
Recipe 3	55.0	41.3	2.0	1.7	0.95
Carbon impact ingredient [kg CO_2-eq/kg]	0	2.13	1.18	2.69	
Source		[46]	[47]	[48]	

5. Discussion

The results in this study lead to the following insights:

- **Waste-filler type:** There were significant variations in the performance of the different food-waste based fillers. Results from the first tranche of testing show that more inert materials (such as walnut shells and spent coffee grounds) were generally better constituents for flawless, strong bio-composites, when compared to more soluble and reactive filler materials (such as cacao bean shells) which resulted in blistering and cracking of the bio-composite.

This blistering, as observed for the variants with CS, RCS and DCG filler, could likely be attributed to the soluble residue in the filler materials, as these three fillers showed the largest weight loss after filtration and the degradation of solubles through heat and pressure of compression moulding has been shown to cause the release of volatile gases in bio-composites in previous studies [53]. The bio-composite specimens with no surface flaws, cracks or blisters, which were also the filler types with low solubility (see Section 4.1), exhibited the highest mechanically performance and the lowest water absorption.

The prior removal of water-soluble elements from the filler, (e.g. in the filtration process of brewing coffee) could therefore help improve the chemical interaction between matrix and filler. Filler/matrix compatibility could also be improved by pre-treatments that enhance dispersion and adhesion, as shown by [2,11]. Adjusting processing parameters (e.g. optimising moulding pressure, temperature, and curing time to reduce blistering) to better accommodate the material could also improve the outcomes.

Table 11
Approximate energy consumption and equivalent carbon impact of processing steps of composite WS_mix_55.

Processing step	Energy consumption [kWh/kg]	CO_2-eq [kg/kg]	Note on estimation	References
Milling filler	0.67 (per kg filler)	0.238 (per kg filler)	~4 min milling per 100 g filler	[49]
Drying filler	0.3 (per kg filler)	0.107 (per kg filler)	Oven duty cycle 20 %, 2 h drying, 3 kW oven, oven fits 4 kg	[50]
Kneading BMC	0.115 (per kg BMC)	0.041 (per kg BMC)	30 min processing time, duty cycle 20 %, 2 l volume, 95°C, ~3 kW	[51]
Moulding BMC	1.0 kWh (per panel)	0.355 (per panel)	15 min pressing, 6 MPa, ~4 kW, 150°C	[52]

Table 12
CO_2-equivalent impact per kilogram of bio-composite WS_mix_55 and compared materials, based on [44].

Material	Primary production [kg CO_2-eq/kg]	Processing [kg CO_2-eq/kg]	Process
WS_mix_55	0.95	0.59	milling, drying, mixing, hot-pressing
Aluminium	9.11	0.36	extrusion, foil rolling
Coated steel	2.01	0.42	extrusion, foil rolling
Ceramic tile	1.68	> 0.0	cutting
Granite	0.70	0.90	grinding
Softwood (fir)	0.27	0.04	coarse machining

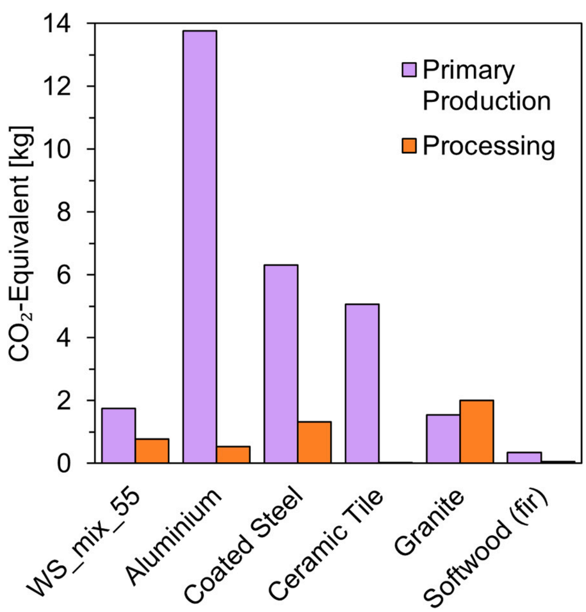

Fig. 14. Equivalent emissions from primary production and processing per panel.

This sensitivity to soluble compounds is particularly evident in the two coffee types resulting in very different bio-composites, even though their material composition and structure are very similar. The de-oiled coffee grounds filler performed worse mechanically than the regular dried spent coffee grounds. The large difference in solubility (9.0 wt% filtration loss for SCG and 23.9 wt% for DCG) likely stems from the DCG material supplier using both spent and fresh coffee grounds for the de-oiling process. With this context, there was no other observable benefit to the de-oiling of coffee grounds, opposed to spent coffee grounds. This disagrees with the findings of We et al., 2016 [54] were interfacial bonding with a polypropylene (PP) matrix was improved through oil extraction.

The weight reduction observed in CS, RCS, and DCG specimens during the water absorption test, after reaching full saturation, could be attributed to the leaching of the soluble compounds into the water bath. This correlates with the faded surface colour of the immersed specimen of these three filler types, even though the colour change could also be attributed to the higher visibility of lighter coloured filler particles, compared to the matrix, on the slightly spalled surface after repeated frost exposure (Fig. 9).

Nonetheless, the furan matrix exhibits good durability and appears to be well suited for high levels of water and frost exposure associated with outdoor applications, even though some changes in surface appearance of the bio-composite are to be expected, as it is also common with other bio-based materials.

The agglomerating behaviour of SCG, DCG, and CP showed that milling and sieving methods need to be customised for different bio-based waste materials.

The observed particle shapes may influence filler dispersion and composite strength. While elongated particles, as observed for WS, could possibly contribute reinforcement effects similar to fibres, rounded particles, as for CP, WS and SCG, could likely support more efficient packing and stress distribution within the matrix [37].

- **Filler particle size:** Results from the second tranche of testing revealed that an increase in grain size leads to lower bending strength for both the WS and the SCG composites, likely due to larger failure-inducing stress concentrations within the matrix and reduced interfacial bonding. This agrees with similar findings reported by others [20,38]. An additional weakness of larger grain sizes is the uneven dispersion of the filler, which can lead to voids and matrix-dominant areas [12,16]. During curing, resin-rich areas may also lead to locally increased crystallisation, resulting in weaker and more brittle areas compared to a uniformly filled composite [12,55].

The blend of grain sizes had contrasting effects on the coffee and walnut fillers. For WS_mix_45, the bending strength exceeded that of the two larger mono-sized walnut filler variants, with a smaller dispersion in strength compared to the variant with filler particles of 125 μm or smaller. Scatter in mechanical data strongly affects design strength values, so low dispersion is desirable for engineering use. SCG_mix_45 on the other hand, performed worse than all other variants of the same filler type in this tranche. These differences could be attributed to the inherent strength of the filler materials or the quality of interfacial bonding. Smaller grains might mitigate poor bonding, while larger grains could lead to agglomerations and weak spots, leading to reduced performance in some filler types as suggested by other studies [12,16].

The inconsistency of scatter with between walnut variants of different grain sizes could not be clearly explained. Repetition of related tests in future work may help determine whether this behaviour reflects an intrinsic material property or a process-related variation.

- **Filler content:** In the third and last tranche of this study the filler/matrix ratio was investigated. For both the WS and SCG composites, the 35 % filler version performed worse than the 45 % filler content, in both bending and impact strength. The mixed-grain size of walnut filler seems to have achieved its intended objective: its performance levels (bending strength and impact resistance) at 55 % filler content were similar to those at 45 % filler content. Even though further experiments are needed to verify, this suggests that grading of particle size distribution leads to a reduction in matrix material without compromising mechanical performance, enabling a reduction of the GWP of the composite ingredients.
- **Carbon footprint:** With a local sourcing of waste material, bio-composites, such as the ones investigated in this study, have a large potential for developing materials with low embodied carbon and with mechanical properties similar to conventional building materials. Since the environmental impact of sourcing waste-based filler materials is generally lower than that of furan resin production, an increased content fillers is effective to lower the bio-composites GWP. Bio-composites from food waste are therefore could be a promising alternative to construction materials commonly used in external applications such as aluminium, steel or stone cladding within the life-cycle stages considered in this study. Compared to a similar façade product, Pretty Plastics tiles made from recycled PVC, which amount to a GWP (biogenic and fossil) of 1,14 kg/kg in A1 and A3 [56], the WS_mix_55 composites with 1,54 kg/kg is slightly higher, without accounting for the positive effect of biogenic carbon sequestration. While these metrics cover only a fraction of a full life-cycle analysis, stages A1 and A3 have been shown to make up the main portion of material related emissions in similar cladding applications [56,57].

6. Conclusion

This study demonstrates that bio-composites incorporating industrial food-waste fillers could provide sufficiently strong low-carbon alternatives to various components in the built environment. However, using waste materials as fillers without pre-treatment can negatively impact bonding and mechanical performance, particularly if the waste materials have a high soluble content. Inert fillers, such as ground but untreated walnut shells, showed the best compatibility with the furan resin matrix, while pretreatment may be needed for other fillers. An increase of grain size from < 125–500 μm lead to a drop in mechanical properties for the tested fillers while strategically varying the grain sizes of the filler material proved effective in reducing resin content for the walnut shell filler without compromising mechanical strength, thereby further increasing the recycled content and reducing the environmental impact of the bio-composite. Other fillers with a high soluble content (e.g. cacao bean shells and unspent coffee) require pretreatment to mitigate the bonding weaknesses and improve mechanical performance.

Future work on bio-composites from food waste should build on the findings of this study while addressing its salient limitations, namely:

- Only a limited selection of waste-based bulk fillers and only the thermoset furan resin were examined; other matrix systems or reinforcement strategies were not explored. Future efforts should therefore focus on optimising filler and matrix combinations, pretreatment methods, and refining particle size distributions for a broader range of bio-waste materials.
- Performance was assessed under short-term bending, impact loads and freeze-thaw cycles; Further investigations are needed to evaluate critical performance parameters such as long-term durability, including UV- and chemical exposure, fire performance, use life and end-of-life scenarios as well as cyclic and long-term loading.
- The use-case calculations were based on mean strength values. A larger number of tested samples could provide sufficient data to determine characteristic strength values for engineering design purposes.
- Future studies should include more detailed characterisation of the fillers and composites, for example hardness and porosity of the filler material, as well as through scanning electron microscopy (SEM), to provide deeper insights into microstructural features and their influence on interfacial bonding and mechanical performance.

To fully integrate these composites into a circular material system, future research should also consider the stability and consistency of food industry waste streams, including seasonal availability, material uniformity, and supply chain logistics for reliable large-scale sourcing. Exploring the impact of upscaling waste-based bio-composite production on the involved value chains in regard to environmental and economic impact could further establish use cases for bio-composites, incorporating food waste.

CRediT authorship contribution statement

Lara Neuhaus: Writing – original draft, Visualization, Validation, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Olga Ioannou:** Writing – review & editing, Supervision, Conceptualization. **Mauro Overend:** Writing – review & editing, Supervision, Conceptualization.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Lara Neuhaus reports equipment, drugs, or supplies was provided by NPSP BV. If there are other authors, they 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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Data Availability

No data was used for the research described in the article.

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