SPENT COFFEE GROUND AS FILLER FOR FIBRE REINFORCED COMPOSITES MANUFACTURED IN A DIRECT BULK MOULDING COMPOUND PROCESS

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Abstract

Being the second most traded good in the world, coffee and particularly its waste products are globally accessible. While several efforts to reuse and recycle spent coffee grounds have been made, they are not yet used as fillers in fibre reinforced composites for secondary structures like shells and covers. The contribution presented here combines the recycling of spent coffee ground with the recycling of dry carbon fibre fabric waste. Applying a direct bulk moulding compound process, dried spent coffee ground can be readily mixed with resin and then processed with long fibres to produce composite plates and structures in a short-time and economic way. It has been shown that spent coffee ground can compete with conventional, inorganic fillers and also improves the ecological footprint by recycling a commonly occurring waste material while keeping specific properties on the same level of bulk moulding and sheet moulding compounds produced from virgin materials. The versatility of the filler allows combining it with the most commonly used fibres, glass and carbon. In addition, polymer products containing spent coffee ground exhibit a distinguished aesthetic and haptic appearance.

1. Introduction

Given today's trend to decrease the weight and increase the mileage of cars, composite materials become more and more established in car design. However, recyclability is a major downside of composites as recycling routes are very complex and thus expensive. Furthermore, large amount of waste occur during production, as up to 30 % of the final component weight may arise as waste during the manufacturing of carbon fibre reinforced plastics. Among this waste are cut-offs of carbon fibre fabric [1, 2]. When complex geometries are cut from rectangular fabrics, the edges remain unused. As carbon fibres and their deposition are costly, recycling can be used to regain valuable raw materials. The aforementioned dry waste is beneficial in terms of recycling. The material has not yet reacted with chemical components such as the resin and thus less process steps are needed to recover a high-quality raw material. Comprehensive analyses of the current status of the recycling of carbon fibre composites can be found in various studies [3–7].
Regarding the direct reclamation and reuse of carbon fibre production waste, the authors, together with Saburow et al. [8, 9] have presented a direct route: They have demonstrated the applicability and versatility of the direct bulk moulding compound (BMC) process when dealing with recycled technical fibers. Given the characteristics of this process, the fibres cannot be aligned. However, the direct BMC process shows its advantages in terms of cost and time: Discarded carbon fibres only need to be cut into smaller pieces, so-called patches, but no further steps of reprocessing are needed. During the process, the fibres

are not shortened and thus long fibres (circa 50 mm) will be present in the final structure, allowing for high stiffness and strength. While previous works by the authors and by Saburow et al. [8, 9] have focussed on un-filled composites, this work discusses the combination of carbon fibre fabric waste with the recycled filler spent coffee ground.

As coffee is among the worlds most traded goods, no less then 9.5 million tons of coffee are produced and consumed per year [10]. As only a vanishingly low fraction of solids end up in a cup of coffee, most of the ground coffee is disposed of waste. About 50% of the annual coffee production is processed as instant coffee, which would facilitate collecting large amounts of coffee waste for recycling purposes. Despite the simplicity to collect coffee waste, only a minor fraction is recycled [11–13].

Fillers are frequently used in long-fibre reinforced polymers as they offer many benefits. They reduce the total cost of composites, they can reduce chemical shrinkage and thus provide better surface qualities and they can change the appearance polymers in terms of colour and surface feel. Fillers might be anorganic (for instance chalk) or organic in nature (for instance wood powder) and show a variety of shapes from spheric to acicular [14]. Spent coffee ground is qualified in respect to all of these aspects and has thus been used as filler before:

E. Ligo holds a patent of a material containing 32% of SCG in phenol resin to replace other fillers like wood flour by SCG. Although he mentions the general applicability, he does not use SCG in fibre reinforced polymers [15].

Tan et al. introduced a polyester containing 5.5% of SCG. They concluded that smaller grain size of the filler leads to higher tensile strength while stiffness increased for lager particles. By chemical treatments of the SCG, tensile strength could be further increased. These treatments included an extraction of oil from the coffee and a mercerisation with sodium hydroxide to reduce oily substances [16].

Baek et al. used SCG in polylactide matrix and inferred that using methylendiphenylisocyanat as additive had a positive effect in terms of tensile strength [17].

Spent coffee grounds have not only been used as filler as they can be used in various ways instead of dumping it on landfill sites. Coffee and its waste can serve as fertilizer in agricultural systems. Spent coffee ground can be used for heat and or electricity production. It can either be burned directly or coffee oil is extracted and refined to alternate fuels [18, 19]. From a material perspective, spent coffee grounds are nowadays mainly used in textile industry and design [1] as the aforementioned approaches to use it as filler are not commercialised yet. Singtex, a company from Taiwan, uses textile fibres made of SCG and polyester to produce fabrics, especially sportswear [2].

In the work at hand, spent coffee ground (SCG) and carbon fibre fabric waste are used to produce fibre reinforced composites. As described earlier, both materials appear as by-products in large amounts in today's industrial processes and are thus qualified to be recycled in a common product.

2. Materials and methods

Within this work, new glass fibres are compared to carbon fibres regained from production waste. The focus lies upon the recycling material, carbon fibres. However, it is expected that these recycled carbon fibres can compete with new glass fibres when costs and weight are both considered. Furthermore, the conventional industrial filler calcium carbonate (chalk) is compared with spent coffee ground. Mechanical properties are presented and physical properties are briefly discussed.

2.1. Material

This work targets a holistic approach of material reuse by combining carbon fibre fabric waste with spent coffee grounds. These materials are compared with commonly used materials: Chalk as filler and glass fibre as reinforcement. The different combinations are displayed in figure 1, which also presents photographs of the fibres and fillers used within this study.

[1] https://www.kaffeeform.com/de/contact
[2] http://www.singtex.com/en-global/home

Fibres The fibres used in this study were of the following types:

- Bi-directional, non-crimp carbon fibre fabric was cut to square patches of 50 × 50 mm. The fabric was of type *PANEX35* by *Zoltek*. The fabric contained polyester sewing yarn and showed a total area weight of the fabric was 300 g m^{-2}. The sewing yarn was not removed before the BMC kneading process. This patch material corresponds to waste material gained in industrial preforming processes.

- Glass fibres were chopped using sheet moulding compound machinery to a length of 1" (25.4 mm). Glass fibers were of type *Multistar 272* by *Johns Manville*.

Resin The resin for all mixtures was a *Daron ZW 14142* hybrid resin consisting of unsaturated polyester and polyurethane, provided by *Aliancys*. All additives needed for BMC production were added immediately before the kneading process.

Filler The conventional filler calcium carbonate (chalk) was compared to spent coffee ground.

- Calcium carbonate filler was of type *Millicarb-OG* provided by *Omya GmbH*, according to the data sheet showing a mean particle size of 2.7 µm.

- Spent coffee ground came in two varieties: As received after brewing coffee in a standard fully automated coffee machine (labelled *scga*) and after undergoing an additional grinding step (labelled *scgf*). Both types were dried in a vacuum chamber for 5 hours at 105 °C. Type scga was sieved with a mesh opening of 1 mm and the drying was continued for another 5 hours with the aforementioned parameters. Type scgf was ground in a pebble mill for 7 hours and dried analogous to type scga.

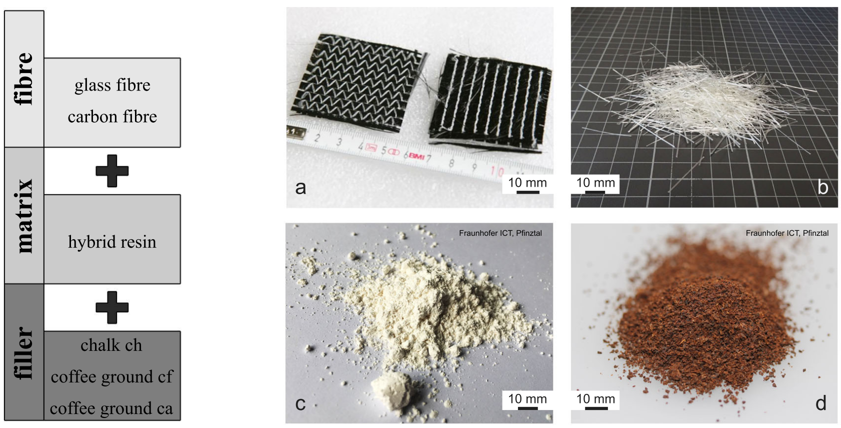

Figure 1. Left: Scheme of material combinations used in this study. Right: Raw materials used in this study. Upper row: reinforcement a) Carbon fibre production waste, cut to square patches; b) chopped glass fibres; Lower row: filler c) chalk; d) spent coffee ground, dried

2.2. Production of BMC

All composite materials investigated in this work were produced in a bulk moulding compound process which will be briefly discussed in the following. For more details about the process route, the reader may refer to earlier works by the authors and by Saburow et. al. [8, 9].

The raw materials were assembled by weight according to the desired formula. Hereby, the filler-resin-paste was filled in the kneader first and fibres were added gradually to prevent machine failure. A homogeneous mixture was achieved after 15 to 20 minutes and the bulk material was portioned and filled in gas-tight bags. The bulks were then stored for 7 days at 20 °C. After this maturation step, bulks were placed in the center of a hydraulic press of type *DYL630/500* by *Dieffenbacher*, covering approximately a ninth of the area. The press was equipped with polished chrome-plated moulds and heated to circa 145 °C. A force of 3200 kN was applied for 110 s. Bulk weight and press parameters were optimised in order to achieve homogeneous BMC plates with a thickness of 3 mm.

From these plates, specimens for different mechanical and physical tests were cut by a CNC water-jet cutting system according to the geometries given in figure 2.

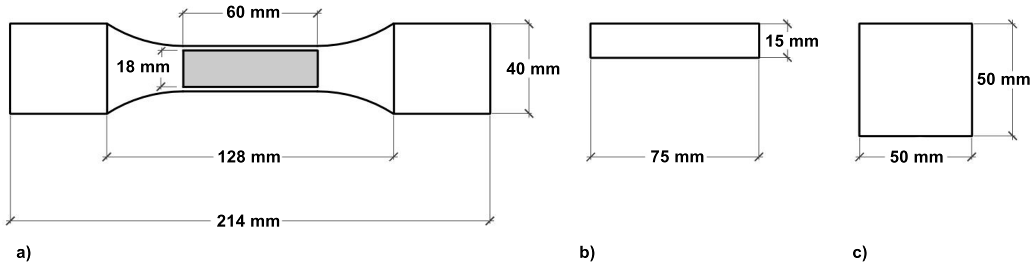

Figure 2. Specimen geometries for tensile (a), bending (b), Charpy (b) and water absorption (c) tests. The grey box highlights the region of interest in the optical evaluation (DIC) of the tensile tests.

To identify the different material variations, table 1 lists names and composition of the materials:

Table 1. Constituents of the materials investigated in this work

specimen ID	fibre		filler		
	CF carbon fibre	GF glass fibre	ch chalk	scgf coffee ground fine	scga coffee ground as received
	in weight-%		in weight-%		
GF_ch	30.0	–	37.8	–	–
GF_scgf	25.0	–	–	24.9	–
GF_scga	25.0	–	–	–	24.9
CF_ch	–	20.0	42.7	–	–
CF_scgf	–	20.0	–	26.5	–
CF_scga	–	20.0	–	–	26.5

According to the material formulas and process stated above, plates were moulded and a selection of specimens is presented in figure 3. As can be seen, the materials containing spend coffee ground as filler exhibit a wood-like, natural appearance.

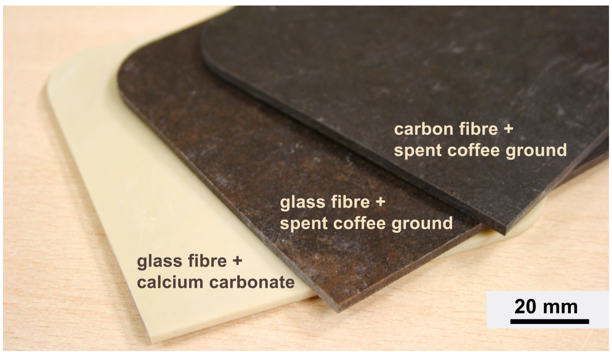

Figure 3. Selection of materials produced for this study

2.3. Test procedures

Tensile tests were run on a *Zwick/Roell Zmart.Pro* universal testing machine equipped with a 200 kN load cell. Specimens were gripped with hydraulic clamps applying a pressure of 20 bar. The initial load was set to 20 N and the test speed to 1.8 mm/ min. Testing procedure and specimen geometry were based on DIN EN ISO 527-4 [20], see also figure 2. Strains were measured using digital image correlation with images taken and processed by a *GOM Aramis 4M* system. Thereby, strains were measured in the region highlighted in figure 2.
Flexural properties were investigated in 4-point-bending mode based on DIN EN ISO 14125 [21] on a *Zwick/Roell Z2.5* device and a load cell with a capacity of 2.5 kN. The initial load was 5 N and the test speed 2 mm/ min.
Impact test were run in accordance with the Charpy procedure based on DIN EN ISO 179-1 [22] on a *Zwick/Roell HIT 5.5P* testing device with a 5J-pendulum, a span of 60 mm and unnotched specimens. Test were run with dry (laboratory conditions) specimens and with specimens that were kept in distilled water for 24 hours at room temperature prior to testing. In this way, it could be observed if the material and particularly the filler, showed intake of moisture and altered impact toughness.
Thermogravimetric analysis (TGA) was run for four specimens for each material and according to a time-temperature-profile recommended by Bücheler et at. Measurements were performed on a *TGA 701*, manufactured by *Leco*.
Density was measured in accordance with DIN 66137-2 [23] on a gaspycnometer of type *PYCNOMATIK ATC* by *POROTEC*.

3. Results

In the following, the physical and mechanical properties and the water absorption of the materials are presented and a short evaluation of spent coffee ground as filler is given.

3.1. Mechanical tests

Results of tensile and flexural tests are presented in figure 4. Displayed in grey are the results for glass fibre reinforced plastics and carbon fibre composites are shown as shaded bars. Chalk-filled materials show slightly higher values in their mechanical properties but the material is also more dense. In regard of lightweight potential, that is tensile or flexural properties divided by the materials weight, chalk- and SCG-filled composites rank on the same level and thus compete with each other. It can further be stated that the additional grinding step for SCG and thus the particle size of the coffee filler is of minor importance.

A closer look on specific properties is taken in figure 5. Exemplary, stiffness determined in tensile tests is divided by the composites density to reveal the specific stiffness. Due to the light filler sgc, the materials containing coffee waste show similar properties as the composites filled with chalk. This effect can particularly be seen in combination with carbon fibres.

As Charpy impact test reveal, the material containing glass fibres and the conventional filler chalk has the superior impact toughness. As can be seen in in figure 5, all other materials rank on the same level of impact toughness as their error bars overlap. No significant influence of the 24 hour soaking of the specimens in water was found.

In addition to the Charpy tests of dry and siw (soaked in water) materials, specimens the size of 50×50 mm (refer to figure 2 c)) were soaked in water for 24 hours and weight prior and after on a high precision scale. Whilst water intake in composites containing chalk was negligible, all composites with coffee as filler showed slight water intake: GF_ch and CF_ch gained less then 0.2% in weight after 24 hours in water. All materials with SCG as filler raised their weight between 0.5% and 0.8% relative to the dried material.

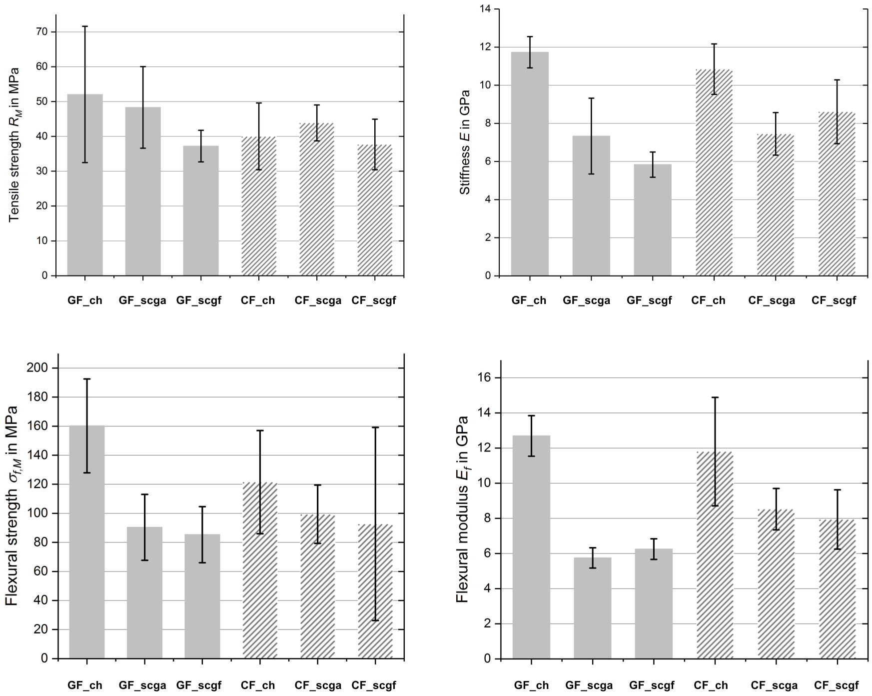

Figure 4. Results for tensile tests (upper row) and bending tests (lower row). Refer to table 1 for informations of material identification and constituents.

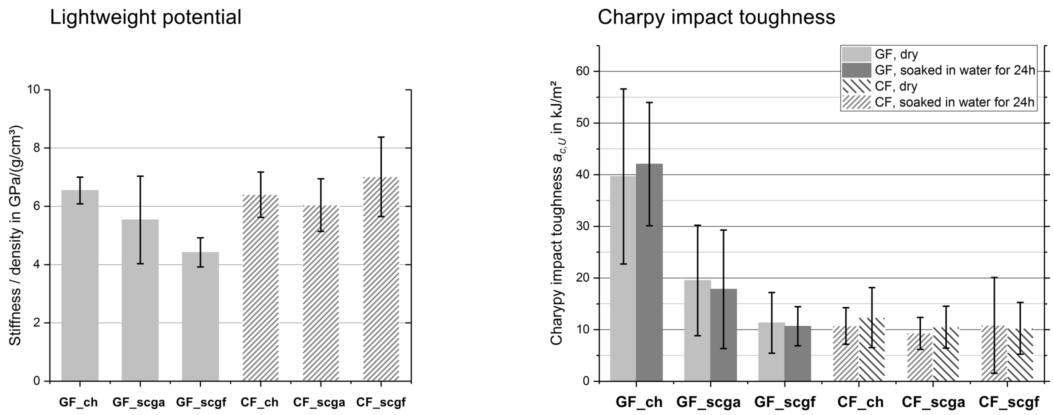

Figure 5. Left: Stiffness normalised with density; Right: Charpy impact toughness

3.2. Determination of fibre content and density

In addition to the mechanical properties, densities and fibre weight content were determined and correlated to the composition as set for the production of the composite materials.

3.2.1. Thermogravimetric analysis

Thermogravimetric analysis (TGA) was used to ensure that the set fibre weight content was hit. Results are listed in table 2. The TGA followed a procedure designed specifically for this type of hybrid resin proposed by Bücheler et at [24].

3.2.2. Density

The density of the conventional filler (chalk) is, according to the data sheet, 2.7 $g\,cm^{-3}$. Density of the coffee filler was measured to be 1.29 $g\,cm^{-3}$ for scgf (fine filler) and 1.31 $g\,cm^{-3}$ for scga (filler as received), respectively. Furthermore, using the data of the composition, the densities of the composites were calculated and are listed in table 2.

Table 2. Fibre weight content (column 2, 3 & 4) and composite density (column 5)

material	set value in weight-%	actual value in weight-%	rel. deviation in %	density composite in $g\,cm^{-3}$
GF_ch	30	31.00	1.78	1.79
GF_scgf	25	25.84	3.34	1.33
GF_scga	25	28.25	12.99	1.32
CF_ch	20	18.29	8.55	1.70
CF_scgf	20	21.11	5.57	1.23
CF_scga	20	21.45	7.24	1.23

4. Discussion

As TGA revealed, the maximum relative deviation of fibre weight content was 13%. Taking this into consideration by dividing the specific stiffness (see figure 5) by the actual fibre weight content, further decreases the difference between chalk and SCG as fillers. Hence, in lightweight applications, SCG can compete with conventional filling materials. As mentioned, fibre bundles are orientated randomly in bulk moulding compounds and thus these materials are locally inhomogeneous (see also [8, 9] and figure 6 b)). Optical strain measurements via digital image correlation during tensile tests and scanning electron micrographs of fracture surfaces, as shown in figure 6, lead to the following conclusion: The composite has a higher risk to fail in those regions where bundles of fibres are orientated perpendicular to the load direction. This characteristic is typical for BMC and SMC materials and cannot be circumvented in composites with a random fibre orientation. Only elaborate and costly intermediate steps of fibre orientation and conditioning are able to avoid this downside of recycled carbon fibres [3, 4]. However, the work at hand aimed at demonstrating a simple process to reuse waste materials and keeping the need for preparation as low as possible. In addition, the fibre matrix interface in the carbon fibre composites was less good then expected. In scanning electron micrographs fibre pull-outs could be found (figure 6 c)). Furthermore, indentations of fibres were found in fracture surfaces, accompanied by fibres without resin residues, indicating a poor interface (6 a and c)). However, this was not due to the coffee or its chemical components as it was also seen in the materials containing chalk and in earlier works of the authors and Saburow et al.[8, 9]. The more likely cause is a faulty linkage between the fibres reactive groups and the hybrid resin.

a) b) c)

Figure 6. Characteristic features of fracture surfaces of BMC materials: a) fibre indentation b) inhomogeneities and random orientation of fibre bundles c) fibre pull-out

Given the mechanical properties gained in this work, it must be concluded that chemical conditioning steps for the SCG would have further increased the mechanical performance of composites filled with SCG.
Besides weight reduction and the good specific properties, the coffee filled fibre composite exhibits distinguished haptic and aesthetic properties as they resemble wood plastic composites.
The mechanical properties in combination with the wood-like appearance qualify the material to be used in interior design for cars and other vehicles. As the material introduced in this work further stands for sustainability and recycling, it can serve as an alternative to conventional composites in future car design.
As positive side effect, odour nuisance during composite production was reduced to the coffees ability to absorb odours. The effect has also been used in clothes containing spent coffee grounds, produced by the company Singtex.

5. Conclusion

In this work it was shown that spent coffee ground can be used as filler in fibre reinforced composites for secondary structures. In regard of specific properties (weight-related), spent coffee ground can compete with the conventional filler calcium carbonate. Down to the present day, spent coffee ground was only used in textile materials and design applications but the work at hand proves its ability to be used in structural application, for instance in interior design for cars. Water absorption of the natural material was uncritical and impact resistance was not reduced due to a slight water intake.
Given its outstanding haptic and aesthetic properties in combination with good mechanical properties, the material is likely to be used by designers who want to use sustainable materials. As shown, the positive environmental impact can be further improved by using recycled fibres.
However, to achieve market maturity, industrial-scale procedures to regain, dehumidify and condition the spent coffee grounds must be developed.

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