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Mechanical and environmental evaluation of ground calcium carbonate ($CaCO_3$) filled polypropylene composites as a sustainable alternative to virgin polypropylene

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ABSTRACT

Polypropylene (PP) has raised environmental concerns particularly for its depletion of fossil-fuels and contribution to climate change. To lower environmental impacts, PP can be combined with biobased fillers such as calcium carbonate ($CaCO_3$). The mechanical and environmental properties of $CaCO_3$ filled PP have not yet been explored in depth. Therefore, this study examines the aesthetic, tensile, flexural, impact, and environmental (via life cycle assessment) properties of injection moulded $CaCO_3$ filled PP with filler content ranging from 0% to 40% at 5% increments. As filler percentage increased, yield strength decreased (0% $CaCO_3$: 17.68 MPa, 40% $CaCO_3$: 12.73 MPa), but young's modulus, flexural modulus, and impact strength increased (respectively 69%, 51%, and 35% greater than pure PP). Flexural strength increased initially at 5% $CaCO_3$ but then declined as more filler was added. A yellowish hue was observed within all blends which growed stronger with more filler. The addition of $CaCO_3$ reduced the environmental impact for all 11 impact categories. For every 5% of $CaCO_3$ added, the material's global warming potential (GWP) decreased by 100g CO_2 eq. per functional unit (1000 cm^3) of composite. Abiotic depletion of fossil fuels declined by 32% when 40% $CaCO_3$ was added. In addition to this study, it would be beneficial to explore other factors that affect the properties of $CaCO_3$ filled PP such as particle size, particle distribution, and binding additives.

1. Introduction

In 2020, 380 million tonnes of plastic were produced globally [1], a number which is growing every year and expected to reach over 1.1 billion tons by 2050 [2]. Polypropylene (PP) is one of the most commonly produced plastics and accounts for 16% of total global plastic production [3]. PP has come under scrutiny for contributing to a variety of environmental issues with depletion of fossil fuels and impact to climate change of particular concern [4]. In order to improve the sustainability of PP, a possible approach is to combine virgin polypropylene with materials deemed more sustainable, such as: rice husk [5,6], Lignin [7,8], Starch [9], Chitosan [10], Talc [11], Polylactic Acid [12], Cellulose [13], Jute [14], Kenaf fibre [6], and Calcium carbonate [15].

Out of the mineral-based fillers, ground calcium carbonate ($CaCO_3$) is the most popular [16] due to its numerous benefits such as being widely available [17], cheap to produce [18], non-toxic [17], and naturally occurring [19]. $CaCO_3$ can be used as a nutrient supplement and FDA approved for use within food and medical applications [20]. $CaCO_3$ is produced from the mining and subsequent grinding of naturally occurring forms of calcium carbonate such as limestone, marble, or chalk [21]. It can regularly be found within polymers such as linear low-density polyethylene [22,23], low-density polyethylene [24,25], high-density polyethylene [26,27], polyvinyl chloride [28–30], polystyrene [31,32], polylactic acid [33,34], and polypropylene [35–38].

$CaCO_3$ has been shown to improve certain mechanical and thermal properties of the resulting polymer composite such as increased thermal resistance [29], impact resistance [39], and elastic resistance [40]. $CaCO_3$ filled polypropylene in particular has been researched for its effect on yield strength [41], young's modulus [42], volume strain [43], tensile strength [44,45], impact toughness [16], flexural properties

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[46], and rheological behaviour [47,48]. It has been found that as filler percentage of $CaCO_3$ within the formulation increases, the potential for issues during processing as well as reduced flexural toughness and strength of the resulting composites could occur [35]. Despite this, the research currently available does not test high percentages of $CaCO_3$ filler whilst increasing at regular increments. Some papers explore up to 40% $CaCO_3$ but at only 10% increments [49–52], whereas others increase by 2–10% increments but only up to 30% [39,53–55]. This limits how accurately manufacturers can extrapolate an ideal percentage of filler to use for their own production requirements.

Studies about samples' appearance once prepared via injection moulding are also lacking. Material manufacturers looking to incorporate $CaCO_3$ into their polypropylene are unable to use scientific literature to decipher how surface appearance of the polypropylene will change as $CaCO_3$ filler is added. Therefore, comparison between colour and aesthetic features of composites containing various concentrations of filler are unable to be made.

In addition to testing mechanical properties, no studies explore the full environmental impact of $CaCO_3$ filled polypropylene composites. This is despite research claiming $CaCO_3$ to be more sustainable based solely on its lack of need for fossil fuels and low embodied energy and carbon footprint [56]. As of 2016, one kg of $CaCO_3$ has a global warming potential (GWP) of 39.6g CO_2 eq [57] (which includes the mining of the limestone and preparation into processable form), compared to one kg of PP which is much greater at 1.95 kg CO_2 eq [58]. Not many other environmental impact categories outside of GWP have been investigated in regard to ground $CaCO_3$. PP, however, has been shown to have a wide range of negative effects on the environment including impact on marine life [59], eutrophication [60], and resource depletion [61] to name a few. The production of the raw virgin material of PP constitutes a vast proportion of its overall impact across its lifecycle [62]. Whether replacing a proportion of the virgin PP with $CaCO_3$ will solve this range of environmental problems is yet to be determined.

To fully understand all environmental impacts, a more in-depth and comprehensive analysis will need to be conducted. Life cycle assessment (LCA) methodology is a well-known and standardised framework to assess the environmental impact of products or services by compiling the input and emissions data throughout the lifecycle to calculate overall environmental impact [63]. As of currently, no environmental assessment study has been conducted on $CaCO_3$ filled polypropylene; a gap this study intends to fill.

2. Materials and methods

The goal of this study is to explore changes in key mechanical, aesthetic, and environmental properties as varying percentages of $CaCO_3$ filler are added to virgin PP to form PP - $CaCO_3$ composites. The environmental impacts across a comprehensive range of impact categories are provided via the use of the life cycle assessment (LCA) methodology.

2.1. Materials

Two commercially available materials were used for this study; Polypropylene (PP) and Ground calcium carbonate ($CaCO_3$). The PP is a polypropylene impact copolymer acquired from the manufacturer INEOS and is manufactured within the United Kingdom (UK). This material has a melt mass flow index (190 °C/21.6 kg) of 20.0g/10 min, tensile yield stress of 21.0 MPa, and Flexural modulus (23 °C) of 850 MPa. A ground calcium carbonate ($CaCO_3$) mineral masterbatch with the product number Granic535 was acquired from a UK-based distributor called Plastribution and originally produced in Spain. This masterbatch has a calcium carbonate content of 83%, with the remaining 17% consisting of a multifunctional copolymer. This material has a melt flow index (190 °C/5 kg) of 1.6g/10 min and a mean particle size (D_{50}) of 2.5 μm.

2.1.1. Injection moulding processing

Both the $CaCO_3$ and polypropylene were purchased in the form of pellets which were introduced into an injection moulding machine via a hopper allowing for distribution of the $CaCO_3$ within the polypropylene. Measured weight percentages of $CaCO_3$ and polypropylene were added to the hopper to create the various composites of varying filler percentages. $CaCO_3$ masterbatch was added to the polypropylene in percentages of 0%–40% at 5% increments (therefore equalling 4.15%–33.2% of pure $CaCO_3$ at 4.15% increments). This results in nine specimens of varying filler percentages being created. An ISO 527 type 1 dumbbell test bar mould with thickness 4 mm and 170 mm in length was used following standard BS EN ISO 527–2:2012 [64].

2.2. Methods

This study applies four methods to assess the mechanical and environmental properties of the composites; tensile tests accompanied with the Young's modulus formula, 3-point bend tests, Charpy impact tests, and Life Cycle Assessment methodology. These methods are described below. Aesthetic properties are assessed using visual observation.

2.2.1. Tensile testing

Tensile tests were carried out according to standard ISO 527–1:2019 [65] using an Instron 5967 tensile test system at a cross-head speed of 10 mm/min using a 30 kN load cell. The samples used were type 1 dumbbell test bars with a thickness of 4 mm and a total length of 170 mm. The average value of three samples for each type of composite was taken. The samples were tested at room temperature (20 °C). All data was recorded by the Instron test system with no need for an extensometer. The yield stress was calculated by identifying the greatest tensile stress within the tensile stress-strain curve data and the young's modulus was calculated using the equation:

$$Young's\ modulus\ (E) = \frac{\text{Stress } (\sigma)}{Strain\ (\varepsilon)}$$

An image of the tensile testing equipment set-up used is provided in Fig. 1.

Fig. 1. Image of the Instron 5967 tensile testing system used for the tensile tests.

2.2.2. Flexural (3-point bend) testing

Flexural properties were measured via a 3-point bend test according to standard ISO 178:2019 [66] using an Instron 5967 test system at a cross-head speed of 10 mm/min. The average value of three samples for each type of composite was taken. The tests were conducted at room temperature (20 °C). The flexural strength and flexural modulus were calculated using the following equations:

Flexural strength: $\sigma_f = \frac{3F_{max}L}{2WT^2}$.

Flexural modulus: $E_f = \frac{L^3m}{4WT^3}$.

- L (span) = 64 mm
- T (thickness) = 4 mm
- W (width) = 10 mm
- F = force applied
- m = gradient of the initial straight-line section of the load deflection curve

A diagram demonstrating the set-up for the flexural test is displayed in Fig. 2.

2.2.3. Impact testing

The Charpy impact testing of the composites was conducted on an Instron CEAST impact tester according to standard ISO 179–1:2023 [67]. a pendulum of 2 J impact energy was used to measure the specimens over a cross sectional area of 70 mm × 10 mm x 4 mm. For each composite, three specimens were tested. The following equation was used to calculate Charpy impact strength:

Charpy impact strength = $\frac{\text{Impact energy}}{\text{Specimen thickness} * \text{Specimen width}}$.

- Thickness = 4 mm
- Width = 10 mm

2.2.4. Life cycle assessment (LCA) methodology

For this study, the environmental impact assessments are performed applying the Life Cycle Assessment methodology according to the ISO standards 14040 and 14044:2006 [68,69], and conducted using the SimaPro software (v8.3.1) [70]. The functional unit was set at 1000 cm^3 (which in this case is equivalent to 1 kg) of composite material with varying percentages of $CaCO_3$ filler (0%–40% at 5% increments). The scope of this assessment is cradle to gate including the following life cycle stages: raw material extraction, transport to pellet processing, pellet production, and subsequent injection moulding to represent required manufacturing into useable parts.

Background information for product flows were sourced from the Ecoinvent 3.2 database [71] with global market averages used. For the life cycle inventory of the raw materials, unprocessed limestone and granulate polypropylene were selected which are detailed in table A1 of the supplementary information (SI). The rest of the life cycle inventory (e.g., transport vehicle, transport distance, injection moulding process, and electricity grid used) can be found in table A2 of the SI. The transport and pellet production impacts are embedded within the raw material extraction process as provided by the Ecoinvent database. The Life Cycle Impact assessment results were calculated using the CML-IA Baseline version 3.03 EU25 methodology including all 11 environmental impact categories.

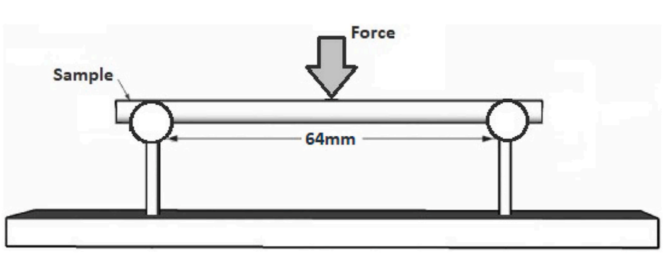

Fig. 2. Diagram demonstrating the set-up for the 3-point bend flexural test.

3. Experimental results and discussion

The results from each test are provided and discussed below in separated sections. The aesthetic properties are discussed in section 3.1., tensile properties in section 3.2., flexural properties in section 3.3., impact properties in section 3.4., and environmental properties in section 3.5. An overall discussion of these properties combined are then discussed in section 3.6. followed by suggestions for future research in section 4.

3.1. Aesthetic properties

Fig. 3 displays images taken of the tensile test pieces created via injection moulding for each formulation. The 0% specimen is shown to have an opaque white colour and even just 5% of filler added creates a noticeable difference in surface colour. 5% filler shows to be a very light cream which becomes only a slightly darker cream at 10% and 15%. A yellowish hue can be seen to become more prevalent as the percentage of $CaCO_3$ filler increases. At 20% filler and above, the yellowish tint appears to no longer grow much stronger but instead maintains a similar dark cream colour. The colour change across all specimens is uniform in that the colour is equally distributed for each piece. There are no noticeable areas of irregular concentration of colour gradients within individual samples. This indicates that there is minimal clumping of $CaCO_3$ particles on a macro scale allowing for the colour to spread evenly.

When the specimens were moulded, no alterations to the injection moulding equipment were required compared to the requirements when moulding pure polypropylene (0% $CaCO_3$). No mixing of the polypropylene pellets and ground calcium carbonate was conducted prior to being placed in the machine but via observation of colour, generalised distribution of materials appears to have occurred. In addition to colour, no observable changes to the macro scale topography were noted. The surface of the composites where $CaCO_3$ particles were added did not appear to have any noticeable roughness or uneven texture compared to the sample of pure polypropylene (0% $CaCO_3$).

3.2. Tensile testing

Fig. 4 shows the tensile stress-strain curves for the polypropylene - $CaCO_3$ (0%–40%) composites. Accompanied alongside the stress-strain

Fig. 3. Visual appearance of $CaCO_3$ - polypropylene composites with 5% increments of $CaCO_3$ filler (From left to right, CaCO3 content ranges from 0% to 40%, increasing at 5% increments).

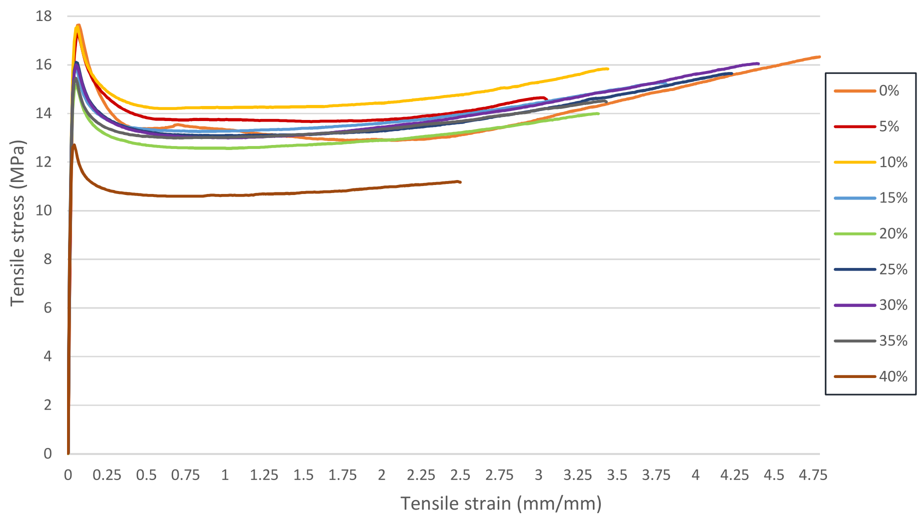

Fig. 4. Tensile stress-strain curves of polypropylene composites filled with calcium carbonate ($CaCO_3$) ranging from 0% to 40% at 5% increments.

curves are the yield strength values and young's modulus of the specimens provided in Table 1. To aid the reader, a magnified version of the tensile stress-strain curves from 0 mm/mm to 0.25 mm/mm strain is provided in Fig. 5. As three tests were conducted for each composite, the test which provided the smoothest curve for comparison has been used for the graphs.

The stress-strain curves show how all specimens demonstrate tensile behaviour typical of plastic ductile yielding followed by eventual fracture. The specimen containing 0% $CaCO_3$ is shown to have the largest elongation at break, fracturing at a strain of 4.79 mm/mm. The 40% $CaCO_3$ filled polypropylene sample has the lowest elongation at break by a large margin and seen to fracture at 2.5 mm/mm strain, much less than the other specimens and around half of the elongation of pure polypropylene. The other samples are shown to break at values between these two formulations. 30% $CaCO_3$ filled polypropylene has an elongation at break closest to that of pure polypropylene (0% $CaCO_3$) breaking at a strain of 4.40 mm/mm; 0.39 mm/mm less than that of pure polypropylene. It appears that as $CaCO_3$ filler content increases from 5% to 30%, the elongation before breaking increases but then any filler percentage higher, the elongation experiences rapid decrease. At percentages above 30%, breakage is shown to occur at 3.43 mm/mm and 2.5 mm/mm for 35% and 40% respectively. This is a fast decline from the break of 4.40 mm/mm when using 30% $CaCO_3$.

Table 1
Yield strength and Young's modulus of polypropylene composites filled with calcium carbonate ($CaCO_3$) ranging from 0% to 40% at 5% increments.

Percentage $CaCO_3$ filler	Yield strength (MPa)	Yield strength standard deviation (MPa)	Young's modulus (MPa)	Youngs modulus standard deviation (MPa)
0%	17.68	1.2	733	72.3
5%	17.26	0.5	818	23.4
10%	17.60	0.7	925	47.5
15%	15.86	0.1	942	55.5
20%	15.17	1.0	961	80.8
25%	16.14	0.4	1009	29.2
30%	16.02	1.2	1168	78.9
35%	15.46	1.0	1205	75.2
40%	12.73	0.2	1240	28.9

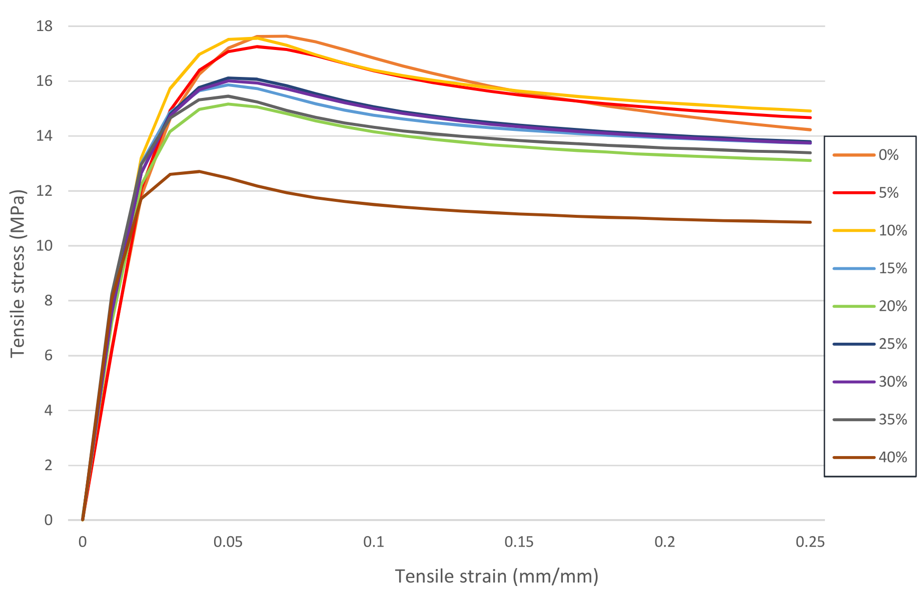

Fig. 5. Magnified version of the tensile stress-strain curves of polypropylene composites filled with calcium carbonate ($CaCO_3$) ranging from 0% to 40% at 5% increments from 0 mm/mm to 0.25 mm/mm tensile strain.

It can be seen in both Fig. 4 and Table 1 that as percentage of $CaCO_3$ present increases, the yield strength reduces. This trend is not perfectly linear but does show a steady overall decrease. Pure polypropylene (0% $CaCO_3$) has the greatest yield strength of 17.68 MPa but is only slightly greater than that of 5% and 10% (17.26 MPa and 17.60 MPa respectively). At 15% $CaCO_3$ filler and above, the yield strength demonstrates a larger decrease. For 15% and 20%, yield strength lowers to 15.86 MPa and 15.17 MPa respectively, a 10% and 14% reduction from pure polypropylene (0% $CaCO_3$). Interestingly, at 25% and 30%, the yield strength increases slightly to 16.14 MPa and 16.02 MPa. This is still over 1.5 MPa lower than pure polypropylene but higher than the composites containing lower percentages of filler (15% and 20% $CaCO_3$).

[51] and [49] studied yield strength up to 30% $CaCO_3$ at 10% increasing filler percentage increments. They both found that yield strength decreases consistently with no percentages where the value increased as higher filler content was added. This study used an average of three samples when calculating tensile properties, all samples of which demonstrated the increased yield strength for 25% and 30% $CaCO_3$ filler. This indicates that another factor is at play which differs between this study and the previous two resulting in the increased yield strength of these specimens. Perhaps at these percentages, changes have occurred between the interaction between the $CaCO_3$ particles and the polypropylene but with current understanding, this will need further investigation.

Just as was seen with elongation at break, when $CaCO_3$ filler percentage is at 25% or 30%, yield strength is closer to pure polypropylene (0% $CaCO_3$) than specimens containing lower percentages of $CaCO_3$ filler (15% and 20%). Also as was seen with elongation at break, yield strength decreases significantly and is the lowest (12.73 MPa) for the sample containing 40% $CaCO_3$. This sudden drop in both elongation at break and yield strength indicates that at this filler percentage and above, the material will have noticeably different tensile properties to pure polypropylene and may not be a suitable material alternative.

Table 1 shows that as $CaCO_3$ filler percentage increases, the young's modulus increases. The pure polypropylene (0% $CaCO_3$) sample has the lowest young's modulus of 733 MPa, whereas 40% $CaCO_3$ filled polypropylene has the highest at 1240 MPa; a 69% increase. Between each increment of +5% $CaCO_3$, the young's modulus can be seen to increase consistently with each addition. As young's modulus measures the stiffness of a material, these results show that as percentage of $CaCO_3$ filler increases, the stiffness of the material also increases.

It is important to note that with the standard deviation taken into consideration, despite the average young's modulus increasing, some of the samples with higher $CaCO_3$ content had lower moduli than its preceding samples. For example, looking at Table 1, the difference in young's modulus between the samples containing 10% and 15% filler only varies by 17 MPa but have standard deviations of 47.5 and 55.5 MPa. This indicates that even though the overall average trend of the young's modulus is moving upwards, the exact measurements between each sample can vary to the point where one sample with a higher percentage of filler has a lower young's modulus than a sample with less filler. A previous study theorised that the cause of this occurrence is due to the consistency of the filler particle dispersion and distribution within the samples [72]. Despite a sample having more filler present, if the agglomeration of the particles is not consistent, this can have slight affect on the overall young's modulus. This results in some samples having higher or lower values compared to samples of slightly different filler contents.

3.3. Flexural (3-point bend) testing

Fig. 6 displays the extension-load curves resulting from 3-point bend flexural tests of the polypropylene composites filled with various percentages of calcium carbonate ($CaCO_3$). These tests were conducted up to 25 mm extension as at this point, the samples showed no signs of breaking and instead started to deform. The key information about the flexural properties of the samples can be demonstrated using the data provided prior to the maximum extension. Accompanied alongside the extension-load curves are the flexural strengths of the specimens and maximum loads (F_{max}) experienced; provided in Table 2.

As can be seen in Fig. 6, the 3-point bend tests were continued until 25 mm extension at which point none of the specimens had fractured. The pure polypropylene (0% $CaCO_3$) sample had a flexural strength and maximum force that was at the midpoint of all the samples. Once 5% of $CaCO_3$ was added, the flexural properties increased dramatically. The

Table 2
Flexural strength and maximum load (F_{max}) of polypropylene composites filled with calcium carbonate ($CaCO_3$) ranging from 0% to 40% at 5% increments.

Percentage $CaCO_3$ filler (%)	F_{max} (N)	Flexural strength (N/mm^2)	Flexural strength standard deviation (N/mm^2)
0	37.55	24.64	0.40
5	46.80	30.71	0.67
10	44.51	29.21	1.01
15	41.28	27.09	1.09
20	36.87	24.20	1.55
25	36.58	24.01	0.97
30	30.73	20.16	0.31
35	31.73	20.82	1.23
40	30.87	20.26	0.67

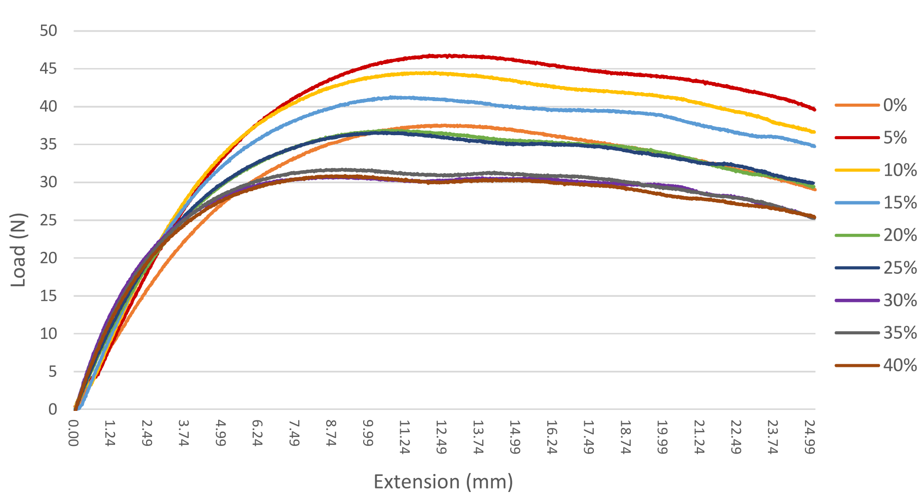

Fig. 6. Flexural extension-load curves resulting from 3-point bend tests of polypropylene composites filled with calcium carbonate ($CaCO_3$) ranging from 0% to 40% at 5% increments.

specimen with the greatest flexural strength (30.71 N/mm^2) and maximum load (46.80 N) was the 5% $CaCO_3$ filled polypropylene. The composites containing 5%, 10%, and 15% $CaCO_3$ filler had very good performance in terms of flexural strength with values higher than that of pure polypropylene. This shows that incorporating small amounts of $CaCO_3$ can positively affect the material's ability to resist deformation. As higher percentages of $CaCO_3$ were added, the flexural strength and F_{max} decreased. The lowest flexural strength can be seen for the 30% $CaCO_3$ filled polypropylene samples with a flexural strength of 20.16 N/mm^2 and maximum load of 30.73 N. Both of these values are 34% lower than the 5% $CaCO_3$ sample and 18% lower than the pure polypropylene (0% $CaCO_3$). A similar study which explored the flexural strength of $CaCO_3$ filled polypropylene up to 25% filler supports these findings as it also found that initial addition of small amounts of $CaCO_3$ increased strength which then gradually decreased as more filler was incorporated [73].

A previous paper suggests that this initial increase in flexural strength at small percentages (0%–15%) of added $CaCO_3$ is due to the toughening effect that calcium carbonate has on polymers when the filler is present at low quantities [74]. $CaCO_3$ has been shown to promote nucleation within polypropylene resulting in enhanced crystallisation and generation of fine sphaerocrystal [73,75]. These improved crystalline properties are then what toughens the polypropylene improving the flexural strength [74]. However as the percentage of $CaCO_3$ filler exceeds 15%, the increased density of filler particles results in agglomeration which impedes uniform distribution and dispersion of filler within the polymer matrix. This leads to reduced stability of the composite and lowers the flexural strength [74].

As flexural strength refers to a material's ability to resist deformation, these results indicate that adding small percentages of $CaCO_3$ will make polypropylene less likely to deform. It is only until above 15% of $CaCO_3$ is added before the flexural strength and maximum force of the composite becomes lower than that of pure polypropylene. As filler percentages reach higher levels (30% and above), the flexural strength and F_{max} appears to stay consistent or even increases as is the case of 35% $CaCO_3$ compared to 30% $CaCO_3$. This is represented quite well in Fig. 6, where the 30%, 35%, and 40% $CaCO_3$ filled polypropylene samples have extension-load curve that stay consistently close throughout the 3-point bend test. This may indicate that at $CaCO_3$ filler percentages of 30% and above, flexural strength and maximum load no longer continues to reduce. Further tests could be conducted at $CaCO_3$ filler percentages of greater than 40% in future experiments to see if the F_{max} and flexural strength stays constant or changes at much higher filler percentage ranges.

Flexural modulus has also been calculated for each polypropylene – $CaCO_3$ sample and this data is shown in Table 3. Table 3 shows that the pure polypropylene (0% $CaCO_3$) sample has the lowest flexural modulus (856 N/mm^2) out of all of the samples. As $CaCO_3$ filler percentage is increasingly added to the composites, the flexural modulus also increases. The 40% $CaCO_3$ filled polypropylene specimen has the highest flexural modulus of 1289 N/mm^2; 51% higher than pure polypropylene.

Table 3
Flexural modulus of polypropylene composites filled with calcium carbonate ($CaCO_3$) ranging from 0% to 40% at 5% increments.

Percentage $CaCO_3$ filler (%)	Flexural modulus (N/mm^2)	Flexural modulus standard deviation (N/mm^2)
0	856	8.09
5	955	1.59
10	1056	4.06
15	1081	12.81
20	1099	10.52
25	1127	5.36
30	1217	1.39
35	1264	4.50
40	1289	3.00

Flexural modulus measures a materials stiffness during bending meaning these results show that pure polypropylene (0% $CaCO_3$), with the lowest score, has the greatest flexibility of all the samples. As more $CaCO_3$ filler is added to the polypropylene, the resulting composite can be seen to increase in flexural modulus and therefore increase in material stiffness with the 40% $CaCO_3$ sample therefore having the greatest stiffness. It can be seen clearly that as greater percentages of $CaCO_3$ filler are added to polypropylene, the resulting material composites become stiffer and at over 15% filler, has lower deformation resistance than pure polypropylene.

3.4. Impact testing

Table 4 shows the average Charpy impact data taken from three samples of each of the $CaCO_3$ filled polypropylene composites from filler percentages of 0%–40% at 5% increments.

Table 4 shows that Charpy impact strength increases slightly as percentage of $CaCO_3$ filler increases with the sample containing 40% $CaCO_3$ filler having the highest Charpy impact strength of 0.03725 J/mm^2; 35% greater than pure polypropylene (0% $CaCO_3$). This increasing trend, however, is not linear and fluctuations in the impact strength can be observed at different filler percentages. In particular, the 10% and 25% $CaCO_3$ filled specimens which have lower impact strength than pure polypropylene (0% $CaCO_3$). This data suggests that the inclusion of $CaCO_3$ has an inconsistent effect on impact strength. As impact strength is known to be affected by a variety of filler properties (e.g., particle size, particle distribution, matrix – filler binding additives etc.), future research could further help to identify which factors could be key in order to ensure consistent modifications to impact strength.

3.5. Environmental impact assessment

Fig. 7 displays the environmental impacts for the $CaCO_3$ filled polypropylene composites (as filler percentages range from 0% to 40%, increasing at 5% increments) per functional unit of 1000 cm^3 of composite material. All eleven environmental impact categories as provided by the CML-IA Baseline version 3.03 EU25 methodology are included. Black lines are provided to indicate the impact provided from the injection moulding process. The remaining impact above the line includes the raw material extraction, transport to pellet production, and pellet processing. These impacts are combined as within the Ecoinvent database, they are embedded together into one process.

As can be seen in Fig. 7, the environmental impact for all eleven categories decreases as $CaCO_3$ filler percentage increases. For each category, the reduction in impact is linearly proportional to quantity of added filler. For example, for every 5% of $CaCO_3$ added, global warming potential's emissions decreases by 100g CO_2 eq. per 1000 cm^3 of composite material. This is consistent whether you are comparing 0% filler to 5% filler or 35%–40%; the reduction per increment is the same.

The overall environmental impact for all categories can be seen to

Table 4
Average Charpy impact data of three specimens for polypropylene composites filled with calcium carbonate ($CaCO_3$) ranging from 0% to 40% at 5% increments. Standard deviation from each set of three specimens is also provided.

Percentage of $CaCO_3$ filler	Average initial Charpy impact strength (J)	Standard Deviation	Average calculated Charpy impact strength (J/mm^2)
0%	1.10	0.09	0.028
5%	1.25	0.26	0.031
10%	1.07	0.20	0.027
15%	1.37	0.15	0.034
20%	1.23	0.08	0.031
25%	1.09	0.10	0.027
30%	1.35	0.06	0.034
35%	1.31	0.17	0.033
40%	1.49	0.08	0.037

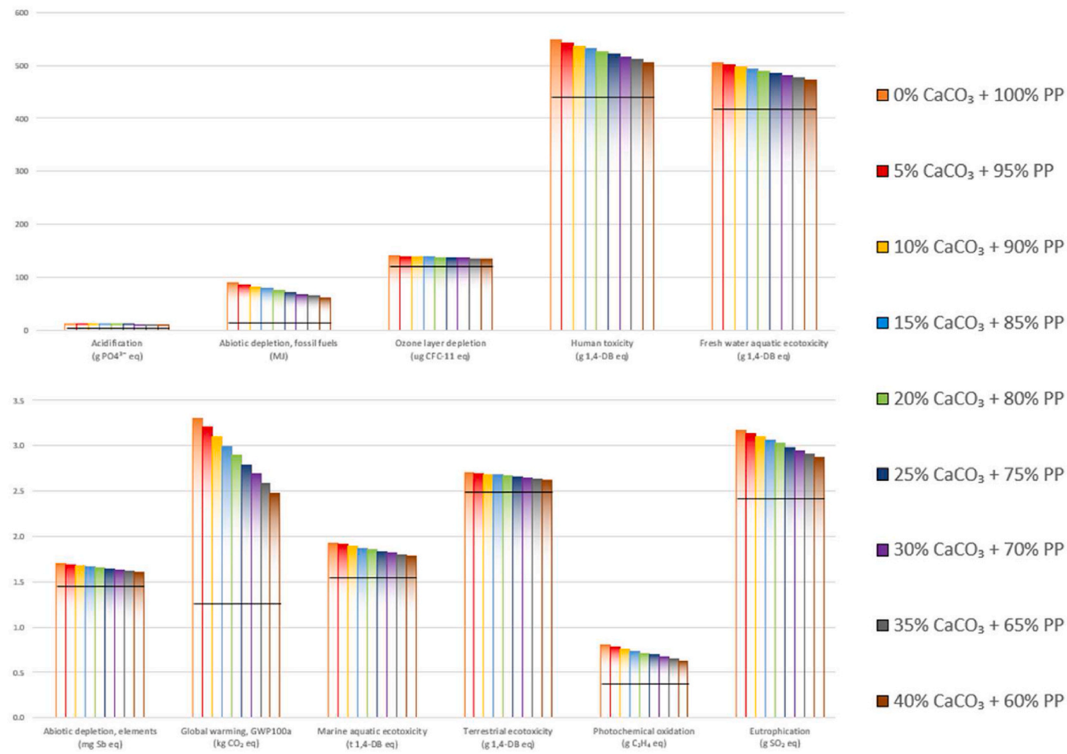

Fig. 7. Graphical representation of environmental impact data for polypropylene composites filled with calcium carbonate ($CaCO_3$) ranging from 0% to 40% at 5% increments. All eleven environmental impact categories as provided by the CML-IA Baseline version 3.03 EU25 methodology are included. Results shown per functional unit of 1000 cm^3 of $CaCO_3$ filled polypropylene. Black lines are included to indicate the impact provided from the injection moulding process.

decrease by 3%–32% once 40% of $CaCO_3$ has been added. The category with the least reduction of impact (3%) is terrestrial ecotoxicity that decreases from 2.7g 1,4-DB eq. emissions from the pure PP (0% $CaCO_3$) sample to 2.6g 1,4-DB eq. from the 40% $CaCO_3$ filled PP. The overall reduction is small because the environmental impact from the injection moulding manufacturing stage consists the majority (92%) of the total score. In fact, the injection moulding has quite a significant contribution for all categories of between 20% and 92%, as can be seen indicated by the black lines.

The greatest reduction is shown for abiotic depletion of fossil fuels which reduces from 89.5 MJ for the pure polypropylene sample to 60.9 MJ for 40% $CaCO_3$ filled polypropylene; 32% lower. It is understandable that abiotic depletion of fossil fuels would have such a great reduction as polypropylene is derived from fossil fuels whereas ground calcium carbonate ($CaCO_3$) is naturally sourced and does not require fossil fuels within the raw materials. The raw material extraction and pellet processing also has a larger overall contribution to this category of 80% resulting in greater overall reductions as $CaCO_3$ is added to replace polypropylene.

There are no environmental impact categories that are made worst by using $CaCO_3$ so no compromises to certain impact categories would be needed to achieve an overall more sustainable material. Even just small additions of $CaCO_3$ filler in replacement of virgin polypropylene would have a positive environmental impact. Manufacturers can therefore use this data to choose what percentage of filler is acceptable in terms of mechanical tolerances and calculate the resulting environmental savings that will be made.

Fig. 7 showed injection moulding to contribute a large proportion of the total environmental impact. It is, however, important to note that studies have shown that using calcium carbonate within polypropylene can improve thermal conductivity of the resulting composite. This enables polypropylene to heat up faster resulting in lower energy requirements during processing [76]. Manufacturers could potentially measure the change in energy requirements when including $CaCO_3$ into their polypropylene. They could also test whether the operation temperature or run time of the injection moulding machine could be reduced whilst still providing materials of satisfactory quality. These energy savings could lead to further reduced environmental impact and future studies may wish to utilise life cycle assessments to examine the reductions in energy requirements and its resulting change in environmental impact as various percentages of $CaCO_3$ filler are used.

3.6. *Overall discussion*

To help aid the comparison of the various properties that have been explored within this study, Fig. 8 provides a heat map indicating which blends demonstrate desirable or undesirable properties ranging from best (dark green) to worst (red).

Overall, it has been shown that adding $CaCO_3$ to polypropylene will have variable effect on the mechanical and environmental properties depending on the percentage of filler added. Environmental impacts are consistently shown to improve as more $CaCO_3$ is added but the same cannot be said for the mechanical properties. In terms of processability and aesthetic properties, no changes to the injection moulding equipment were required resulting in easy manufacturing but a yellowish hue can be seen in the resulting moulded pieces. Even with just 5% $CaCO_3$ added, the colour change is noticeable and gets stronger as more $CaCO_3$ filler is added. If a pure white colour is required by the manufacturer, even the smallest amount of $CaCO_3$ will deem this unachievable.

Fig. 8 shows the tensile and flexural properties transition from greens and yellows to oranges and reds as more $CaCO_3$ is added. This is due to the yield strength and flexural strength decreasing as well as the Young's modulus and flexural modulus rising when percentage of $CaCO_3$ filler increases. Manufacturers would need to determine how low of the yield and flexural strengths and how high of the Young's and flexural moduli would be acceptable for their product in order to decide on the percentage of $CaCO_3$ filler that can be added for their composite. As environmental impact decreases linearly, the more filler that can be added,

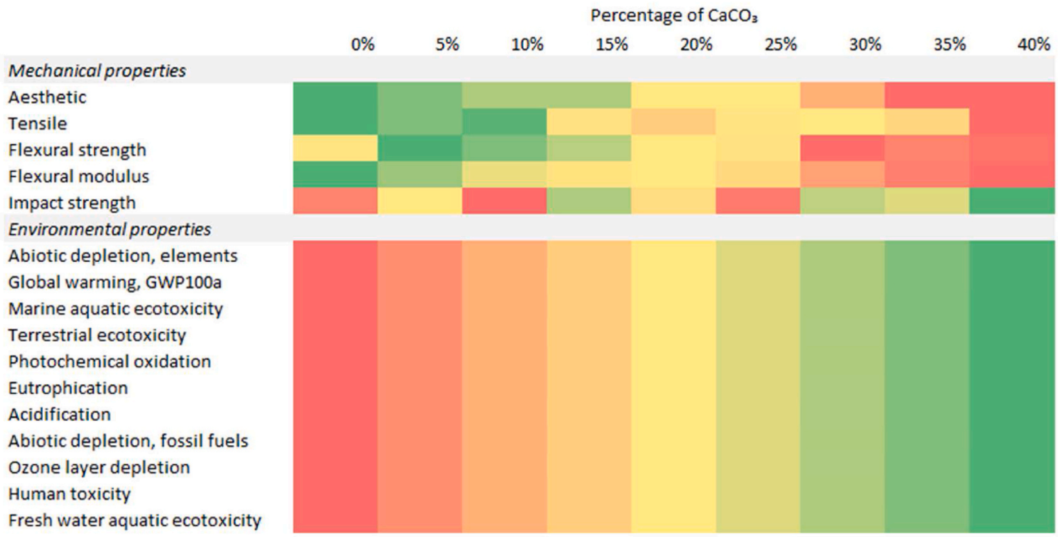

Fig. 8. Heat map showing the performance of various properties displayed by polypropylene composites filled with calcium carbonate ranging from 0% to 40% at 5% increments. The colours indicate the following: Dark green – best, Light green – good, Yellow – midway, Orange – bad, Red – worst. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

the lower the overall impact will be. If a flexural strength closely resembling or surpassing pure polypropylene is desired, then polypropylene containing 20% $CaCO_3$ or less could be used.

Despite the slight overall increase in impact strength as more $CaCO_3$ filler is added, the unpredictable nature of the values as percentage varies makes choosing the best formulation a challenge. This is demonstrated well in Fig. 8 as the colours are shown to not follow any particular pattern and behave sporadically. More research will be needed to explore the effect that different factors, such as particle size, particle distribution, and binding additives etc., may have on the impact strength to determine the cause of this inconsistency.

4. Suggestions for future research

Previous studies have shown that the mechanical properties of composites containing $CaCO_3$ can be dependent on factors pertaining to the shape, size, and dispersion of the $CaCO_3$ particles. Impact strength, in particular, relies on equal dispersion of similarly sized particles within the polymer matrix [77] due to filler particles acting as stress concentrators within the polymer matrix leading to increase fracturing at areas of agglomeration [78]. Section 3.4 of this study showed that impact strength at varying $CaCO_3$ filler percentages can be unpredictable. Electron microscopes could be used to study the particle size distribution within the composite materials and assess how any changes may affect the mechanical properties. This has been done for studies where $CaCO_3$ has been added to polyethylene [79] and when analysing the tensile properties in polypropylene [80].

Changes to the preparation method of the polypropylene - $CaCO_3$ composites could also be explored such as whether premixing prior to injection moulding has any effect on material properties. One study found that a higher back pressure and increased rotation speed of the injection moulding screw can result in superior distribution of filler particles within polypropylene [80]. Variations in injection moulding machine settings could therefore be further tested.

One known property of $CaCO_3$ is its high hydrophilicity which contributes to the agglomeration and uneven dispersion of $CaCO_3$ particles [77]. Calcium carbonate can be treated with compounds of low molecular weight in order to modify how the particles interact within composites [81]. For example, stearic acid decreases the polarity of $CaCO_3$ reducing agglomeration [82,83]. Future research could explore how additives, such as stearic acid, affect the mechanical properties of polypropylene – $CaCO_3$ composites at higher filler percentages.

5. Conclusion

In conclusion, as percentage of $CaCO_3$ filler increases from 0% to 40% within polypropylene, the aesthetic and mechanical properties are affected. In terms of aesthetics, the specimens demonstrate a yellow hue in colour which grows stronger as filler percentage increases. No difficulties were encountered when processing the $CaCO_3$ when using settings usually required to mould pure polypropylene. Elongation at break decreases as filler content increases as well as yield strength which decreases from 17.68 MPa to 12.73 MPa from 0% to 40% $CaCO_3$. The Young's modulus rose consistently as filler percentage increased indicating the materials became stiffer as more $CaCO_3$ was added. Flexural strength initially increased from 24.64 N/mm^2 (pure polypropylene) to 30.71 N/mm^2 as at 5% $CaCO_3$ but then declined as filler increased. The flexural modulus consistently increased as more filler was added; pure polypropylene having the lowest flexural modulus (856 N/mm^2) and 40% $CaCO_3$ filled polypropylene the highest (1289 N/mm^2).

Impact strength increased slightly from pure polypropylene (0.0275 J/mm^2) to 40% $CaCO_3$ (0.03725 J/mm^2), however, this change was inconsistent and unpredictable. Future research is encouraged to explore the affect that dispersion, size, and shape of the $CaCO_3$ particles have on the mechanical properties. The environmental analysis showed that for every category, environmental impact decreases proportionally as $CaCO_3$ filler content increases. For every 5% of $CaCO_3$ added, global warming potential decreased by 100g CO_2 eq. per 1000 cm^3 of composite material. Abiotic depletion of fossil fuels experienced the greatest reduction of 32% when 40% $CaCO_3$ was added.

Overall, this study showed that if the changes in mechanical properties are acceptable to the manufacturers' requirements, that significant environmental savings could be made by partially replacing virgin polypropylene with calcium carbonate ($CaCO_3$). Future research is advised on the effect that particle size distribution, premixing, and binding additives have on the mechanical properties of $CaCO_3$ – PP composites in order to help control some of the properties whilst allowing for the environmental savings associated with using calcium carbonate.

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This research was undertaken whilst receiving a studentship by a UK medical device manufacturer. However, the funding source had no involvement in the design of the study, the collection, analysis, or interpretation of data, or any of the writing stage. None of their background data or intellectual property was used during this study.

CRediT authorship contribution statement

Christina Webb: Writing – original draft, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Kun Qi:** Writing – review & editing, Supervision. **Lorna Anguilano:** Writing – review & editing, Supervision. **Ximena Schmidt Rivera:** Writing – review & editing, Supervision, Project administration, Conceptualization.

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.

Data availability

All the data has been included in the manuscript and SI

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.rinma.2024.100562.

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