# MOULDABLE SMC/BMC AUTOMOTIVE COMPOSITES FROM SUSTAINABLE RESOURCES

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## Abstract

*Recent work at Exeter examined the potential offered by bio-derived fibres, wood and crop-derived particulates, and bio-resins as precursors for SMC. Such materials can deliver a large potential weight saving, as well as offering sustainability credentials. Mechanical performance is however a common shortcoming in such systems compared to GRP, particularly impact behavior. This report details studies into the viability of replacing $CaCO_3$ with a bio-derived particulate filler. The approach taken was to develop a series of SMC formulations using bio-derived fillers. These were then assessed in terms of a) the mechanical performance, b) light-weighting potential and c) sustainability credentials. The importance of controlling paste viscosity is critical for successful processing, fibre wet-out and ultimately composite properties. Different types of bio-derived fillers are shown to impart both thixotropic and dilatent effects, where required viscosities can be "tuned" through blending these of particle types.*

## 1. Introduction.

Short fibre thermoset composites in the form of Sheet and Bulk Moulding Compounds (SMC and BMC) are already used extensively in the automotive industry by virtue of their excellent mechanical /lightweighting properties, low cost and mass manufacturability. SMC represents by far the largest use of thermoset composites in vehicles today, being a lightweight material (1800kg/m$^3$), and therefore much lighter than steel, and challenges aluminum (based on finished part weight). The material therefore, offers much as a technology for LCV of the future. One key negative however surrounds disposability. Impending EU end of life vehicle requirements (ELV) has prompted manufacturers to seek viable routes for disposal at ELV. The SMC class of materials presents environmental and sustainability issues. A recent study at Exeter sought to develop advanced lightweight SMC/BMC grades fabricated as far as possible, from bio-derived organic precursors, including the use of a bio-resin matrix system, organic fillers and natural fibre reinforcement.

In past work concerning SMC/DMC, the focus has been on replacement of just one component, e.g. the use of specific fibres or resin systems in isolation, where very little work has been done on fibre-resin combinations, and none on natural fillers or combined fibre/resin/filler systems. The reason is that previous studies have been prompted by the need for light-weighting or cost reduction rather than disposability. To be truly disposable all three components of the material must be considered rather an any-one component in isolation. This study focuses on bio-fillers, and how they can be utilized in SMC/BMC.

### *1.1 Natural fillers in SMC.*

Wood or cellulose-based particulate fillers have been used in thermosetting plastics since Bakelite, a material based on the thermosetting phenol formaldehyde resin filled with wood flour. The resulting

material was easily mouldable, electrically non-conductive and heat-resistant, and used ubiquitously up to the advent of modern plastics. There has been very little work into the use of natural fillers in SMC/DMC, and there is often overlap between the definitions of the use of natural fibres as either fibre or as fillers; whereby, an increased loading of natural fibres has been simply considered as a filler that reduces the required amount of resin, rather than being an attempt to exploit their reinforcing potential. As simple fillers, the only work where natural materials were used to directly replace calcium carbonate – ubiquitously exploited as a filler in SMC/BMC, involved the use of an organic waste product from sugar production, called bagasse [1]. The BMC developed in this case was designed for the production of injection moulded parts rather than standard compression moulding applications. Bagasse is actually quite a fibrous material that visually looks more like glass fibres than conventional filler particles such as $CaCO_3$. The investigation found that the best mechanical properties were established at very low loadings (25% weight) and were very dependant on the size of the particle used. As with natural fibres there has been more research into the area of simple composites made up of only resin-filler combinations, though in the case of organic fillers the amount of work done on thermoset resins, compared to that on thermoplastics, is even more limited; Chad, 1987 [2] examined polyester resin simply filled with organic waste materials such as rice husk and Marcovich, 1996 [3] with sawdust. Marcovich, 2000 [4] investigated effects of hygroscopicity and dispersion of wood flours used in polyester composites. With the advancement of modern building materials such as fibre boards and reconstituted wood products, there is now, more advanced and specifically engineered technical cellulose based fillers available, (e.g. ARBOCEL® und LIGNOCEL®). These are used in the thermoplastics plastics industry, as functional filler materials in linings, mouldings, seals, insulating mats, rubber wheels, artificial leather covers, hoses. With SMC/DMCs being filled up to 60% with inorganic materials such as calcium carbonate there is great potential to dramatically reduce weight and improve the recyclability of these materials with the use of technical natural fillers. The exploitation potential has not yet been reported at any level, including the production techniques, mechanical properties and compatibility with other ingredients. The overarching targets for this research were as follows:
1) Performance- Compatible mechanical performance to standard parts.
2) Lightweight. - Ideally parts would be less dense than standard parts. Very large potential weight savings through replacement of $CaC0_3$ with bio-derived fillers (The specific density of wood/cellulose is 1400kg/m$^3$ compared to mineral filler with 2700kg/m$^3$.)
3) Disposable. – As far as possible contain bio-derived content and therefore be carbon neutral. – allowing disposal via incineration for energy recovery (leaving only 3% residue), Landfill, Digestion.
In terms of mechanical performance, measurable targets were set in place very early on in the project where new formulations would have to match up to the existing industrially-set performance standards for the particular parts targeted in the project :

1. Non - class A components (e.g. earth mover floor panel) e.g. The JCB floor panel for the JCB backhoe loader.
2. Class A panel (e.g. spoiler & Ford and GOP or grill opening panel for the Ford Transit).
3. BMC application electrical slip-ring part used in the energy industry.

Target performance criteria for each of these parts is described in table1:

| Target Application | MCT (secs) | shrinkage (mm/m) | Density (g/cm3) | Flex Strength (Mpa) | Flex Modulus (GPa) | Impact Resistance (Charpy)(KJ/M2) |
|---|---|---|---|---|---|---|
| **SMC - Floor Panel** | **40-60** | **0.2-1.4** | 1.73-1.79 | 140 | 6 | 50 |
| **SMC - Menzolit Internal Stipulation* for Ford GOP** | **28-38** | **-0.5 to -1.1** | 1.89 - 1.95 | 150 | 8 | 56 |
| **DMC - Slipring stipulation** | **15 -25** | **1.2-2.2** | 1.71-1.81 | 65 | 12 | 10 -Good,<br>20 - Excellent |

**Table 1** Mechanical/Physical Performance Standards of Targeted Parts

**There is no requirement from Ford in standard Class A SMC spec ESB-M3D145-A for MCT, shrinkage or density. The Ford requirement for flexural criteria is somewhat lower, with strength at 125 MPa min, and flex modulus is 7.4 GPa.*

## 2. Experimental details.

Standard SMC and BMC composite production routes were used to produce a range of experimental versions, where the approach taken was to develop a series of BMC/SMC formulations using a) bio-derived resins in place of standard orphothalic unsaturated polyester , b) bio-derived fillers in place of conventional $CaCO_3$, c) combinations of bio-derived resin/filler. A large range of natural fillers were trialed. These were of two distinct types: Cellulosic powders including several wood flour grades, and a range of starches. The resin component used in all Bio-derived SMC/BMC versions developed in this study was DSM's Palapreg® ECO P55-01. The resin is composed of 55% renewable resources.

The resulting panels were then mechanically assessed and compared for their flexural properties and toughness or impact absorption properties, using the Charpy impact test. Such tests are standard mechanical evaluation techniques used in industry for SMC/BMC materials. Results were compared to equivalent glass-reinforced standard grade SMC/BMC, including density comparison for an indication of light-weighting potential, and also an indication of the level of bio-derived content of each formulation.

### *2.1. Materials.*

*2.1.1 Natural Fillers.* Conventional SMC/DMC materials contain up to 60% particulate fillers, usually calcium carbonate. $CaCO_3$ -Particle size typically reported is between 2-5um. There are a number of naturally derived fillers that could make viable replacements for calcium carbonate, bringing advantages in terms of sustainability and light weighting. The programme of research described here embarked on trials with suitable candidate naturally derived powdered filler materials firstly in DMC and then latterly, in SMC formulations in combination with other bio-derived ingredients. The investigation considered a range of cellulosic powders including several wood flours as viable replacements (detailed in table 2). Samples of such materials were supplied by J. Rettenmaier&Söhne GmbH (JRS). Several different types of starch flour were procured and used for 100% $CaCO_3$ replacement in standard SMC/BMC moulding formulations.

| **CELLULOSE FILLER** | **SUPPLIER** | **PARTICLE SIZE** |
|---|---|---|
| Heweten Grade 101- Microcrystalline cellulose | JRS | ~50um |
| Arbocel Grade 100 –Raw Cellulose wood flour | JRS | 70-150um |
| Vitacel Grade 101 powdered cellulose | JRS | 50um |
| Lignocel – grade BK 40-90 wood dust | JRS | 300-500um |
| Arbocel – grade CW-630 PU. Raw cellulose wood flour | JRS | 20-40um |
| **STARCH FILLER** | **SUPPLIER** | **PARTICLE SIZE** |
| Corn flour | MS Baker | Nominal 15um |
| Arrowroot | MS Baker | Nominal 15um |
| Tapioca Starch | MS Baker | Nominal 15um |

Table2. Bio- Filler types considered in this study.

*2.1.2 SMC and BMC pastes*. SMC/BMC are composed of fibre-reinforced paste. The paste is itself, a formulation. Table 3 details the main constituents of the SMC and BMC pastes. All ingredients were provided by Menzolit UK and used as received, where the formulation represents a general purpose SMC/BMC. The exact formulations used are the intellectual property of Menzolit UK and therefore it will not be discussed in detail, However, identical paste formulations was used throughout the study, where only the resin/filler types were altered between batches.

| | SMC | BMC |
|---|---|---|
| Polyester Resin | 1) Derived from maleic acid and standard glycols, dissolved in styrene 2) A saturated polyester in styrene. | Derived from orthophthalic acid and standard glycols, dissolved in styrene |
| Low-Profile Additive | Liquid solution of polyvinyl acetate | Liquid polyvinyl acetate |

| | | |
|---|---|---|
| | (PVCa), dissolved in styrene | (PVCa), dissolved in styrene |
| Inhibitor | p-benzoquinone | p-benzoquinone |
| Initiator | t-BUTYL peroxyester type organic peroxide | t-BUTYL peroxyester type organic peroxide |
| Mould Release | Calcium stearate | Zinc stearate |
| Filler | Calcium carbonate, average particle size of ≤5 µm | Calcium carbonate, average particle size of ≤5µm |
| Reinforcement | Various (Benchmark standard: E-glass fibre rovings cut to 25mm lengths during manufacture) | Various (Benchmark standard: Pre-chopped E-glass fibre rovings of length 6 mm) |
| Thickener | Thickener Magnesium oxide, liquid form | No thickener required |
| Wetting Agent | BYK® W-996 | Not required |

**Table 3** Types of raw materials used for SMC and DMC composite manufacture in this investigation

### *2.2 Specimen manufacture.*

All samples used the same BMC or SMC pastes (as detailed above) where the various fillers examined were compounded at the same volume fraction as the standard glass SMC/BMC used as the benchmark. (SMC 45% vol. fibre, BMC 60% vol.).

### *2.2.1 Standard BMC Compounding.*

All BMC batches were mixed using a 1.5 L ‘Z’-blade mixer. The control BMC batch was prepared using standard processing methods:

1) All the wet ingredients, (resins and catalyst etc), are mixed together for 5 minutes.
2) Then all the remaining dry ingredients except the fibres are added and the paste is mixed for a further 20 minutes.
3) Finally the pre-chopped fibre samples (cut to 6mm) are added and the materials mixed for a final 3.5 minutes (unless otherwise stated).

After compounding the dough was discharged from the mixer and kept refrigerated in a sealed container for 24 hours before moulding.

### *2.2.2 Standard SMC Compounding*

SMC is a well established production method for glass-reinforced thermoset composites. The pre-mixed paste containing all the resin and filler components is fed into the doctor boxes before the production run begins. A constant layer of the paste then covers the carrier films as they are drawn through the doctor box system, and a layer of the particular fibres under test, is then sprinkled randomly onto the bottom carrier film, before it and the top paste-covered carrier film are sandwiched together to form the final SMC mouldable sheets.

The sheets are then stored at 35°C for 48hrs to allow the thickening process to proceed.

SMC sheets, and also BMC dough mixes, are then compression moulded.

### *2.2.3. Moulding*

For each SMC and BMC formulation three flat panels of dimensions 25x25cm and 4mm thick, were moulded, for each of the different formulations, two batches were manufactured separately, to minimise the effect of batch-to-batch variations. Compression moulding was performed with a hot press, where all formulations were cured at 145°C and a pressure of 4.1MPa for 3 minutes, replicating the curing conditions of the standard, industrially produced, SMC/BMC composite.

## 3. Testing.

*3.1 Paste Rheometry measurement.* Capillary Rheometry measurements were used to compare calcium carbonate standard fillers with several natural filler grades (corn 15um and wood flour 40um), to determine and compare the rheological behaviour exhibited by each paste system under similar shear conditions as experienced in so called “high shear” BMC Z blade mixing processes.

The approach taken was to take measurements as close as possible to actual mixing conditions: Temperature, Shear Rate, Shear History. Rotational rheometry proved unsatisfactory –The pastes were unsuitable for flow experiments as the viscosity was too high and pastes fractured rather than flowed. Capillary Rheometry could deliver the required shear rates during flow, and was therefore used for the study.

Samples: 1) $CaCO_3$ Standard BMC paste (no glass), 2) Corn Starch (100% volume equivalent of standard), 3) Arbocel wood flour 100% volume equivalent of standard. All tests were conducted at 30°C, using a Malvern instruments RH10 advanced Capillary Rheometer, where shear viscosity (Pa.s) variation with Shear Rate (/s) plots were obtained for each of the 3 paste samples.

*3.2. Flexural testing.* Three-point flexural testing was conducted in accordance with ISO178 using a Lloyd Instrument EZ20 and a 500 N load cell, with test performed at an extension rate of 1.9 mm/sec. A total of 16 samples were tested for each formulation, with eight samples cut from two of the four cured panels. The sample size used was 80x10x4 mm.

*3.3. Impact testing* Impact energy absorption (also referred to as Impact Strength) was assessed using the Charpy impact test arrangement. Tests were carried out using a Ceast Resil Impactor Junior with a non-instrumented 4 J impact head. Samples were prepared and tested in accordance with ISO179-1 standard. a total of 20+ samples per batch were tested, where samples were taken from several test panels. The sample size used was 80x10x4 mm in line with ISO specification.

## 4. Analysis.

*4.1. Particle size/distribution measurement.* Particulate filler samples were interrogated for particle size distribution using a Micromeritics Saturn DigiSizer particle sizing instrument employing the light scattering analysis technique. CaCO3 (standard) Rice Flour, Corn Flour, Small particle Wheat Starch -as used in the textile industry, Tapioca Starch, Arrow Root, Potato Flour, Wood Flour (Arbosil Grade CW 630 PU).

## 5. Results and Discussion.

*5.1 Particle size analysis and paste Rheology.* Initial trial batches of moulding compound where conventional $CaCO_3$ fillers were replaced with bio-fillers, proved to be problematic. The pastes produced demonstrated a behaviour change when the fibre reinforcement was added. Fibre and wetting and mixing was poor in comparison to conventional pastes, and required extended mixing times – this in turn, resulted in increased fibre damage. This factor, plus the poor fibre wetting resulted in weak composites with low flow characteristics. It was clear from mixing observations that the bio derived fillers have changed the mixing process substantially, where the glass fibre reinforcement remains incompletely mixed. This result was thought to be due to possible rheological differences between the two paste systems, which in turn, could be caused by differences in particle size/size distribution. Figure 1 compares particle size and size distributions for a range of starch or cellulose-based bio-fillers with that of a standard $CaCO_3$ filler grade as used for the production of SMC/BMC pastes.

$CaCO_3$ powders demonstrate an average particle size of 2-8um and shows a wide particle size distribution profile compared to the starches – where there is a far more defined peak size frequency at around 15um for corn starch and 7um for rice. The fine grade Wood flour demonstrates a generally larger particle size with a frequency peak at 45um. Figure 2 compares capillary rheometry traces for several bio- fillers against $CaCO_3$ standards. Interestingly, wood flours were found to shear thin as shear rate increased, demonstrating a classic thixotropic response, whilst corn starches demonstrate dilatants behaviour in fluids, i.e. shear thickening as shear increases. $CaCO_3$ shows a more modest shear thinning behaviour.

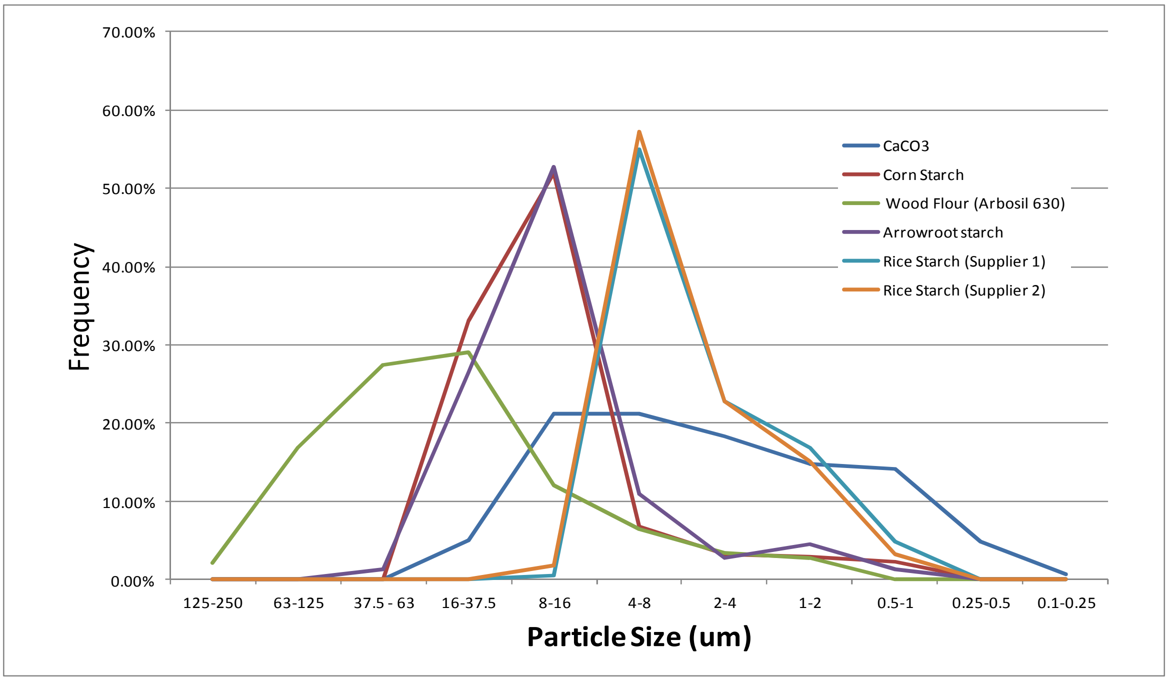

**Figure 1**. Particle size and size distributions for a range of starch or cellulose-based bio-fillers with that of a standard CaCO3 filler grade as used for the production of SMC/BMC pastes.

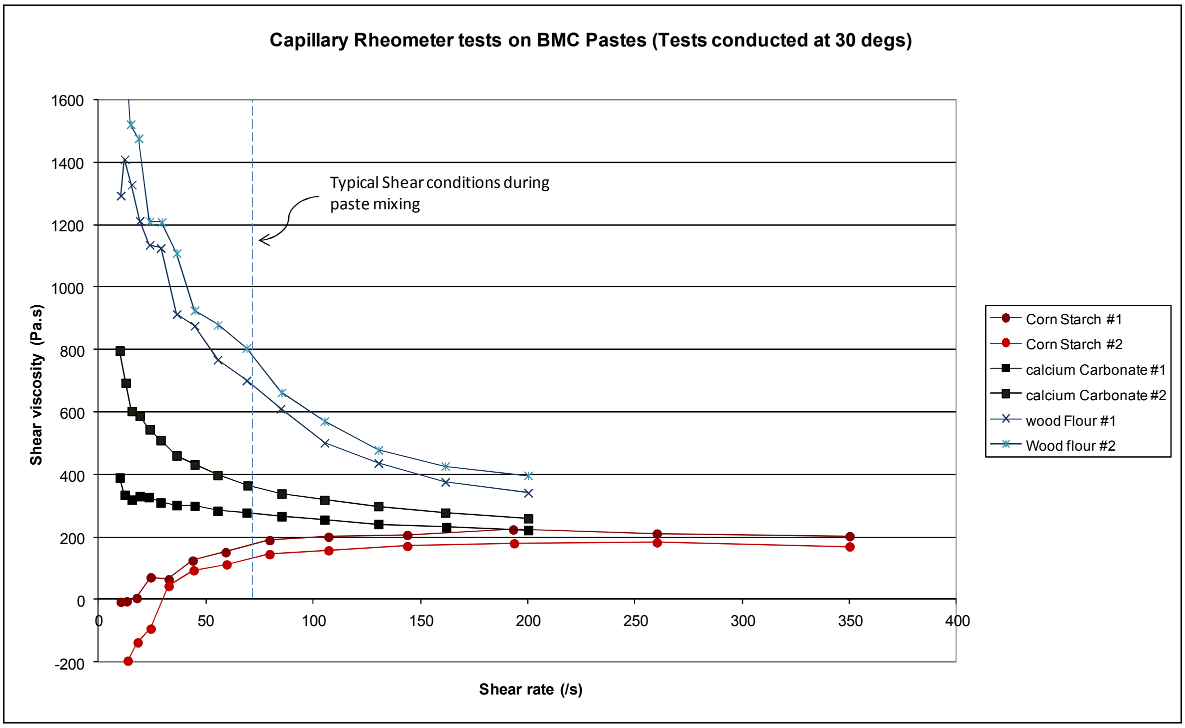

**Figure 2.** Capillary rheometer traces comparing shear viscosity/shear rate for BMC pastes -filled with $CaCO_3$, corn starch, and wood flour. (60% Vf loading).

Figure 2 also indicates the typical shear conditions that exist during the mixing of BMC/SMC pastes. It was realised that it was important that experimental formulations replicate the viscosity behaviour profile of typical moulding compounds, and that this could be achieved by blending the wood flour and starch particles in order to “tune” the paste viscosity to more closely align with typical $CaCO_3$ base formulations.

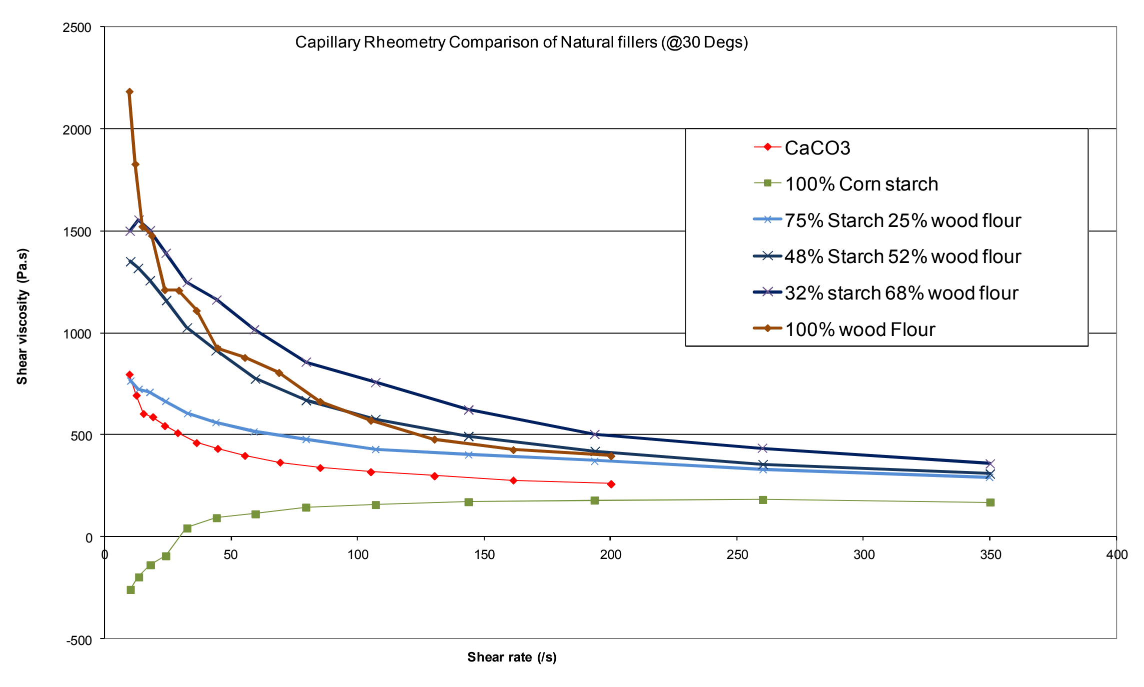

**Figure 3.** Capillary rheometer traces comparing shear viscosity/shear rate for BMC pastes showing the effect of blending on viscosity profiles .

Figure 3 compares the viscosity profiles for a series of blended bio-fillers, where the combined effects of thixotropic wood flours with the dilatent behaviour of starches, can be used to tune paste viscosity. It is immediately clear that the 75% starch/25% wood flour combination, gives a viscosity trace close to that of $CaCO_3$ conventional fillers.

### *5.2 Mechanical Testing.*

Table 4 summarises the mechanical testing results for the range of bio-derived SMC and BMC grades produced in this study. The table gives summarised data for all experimental formulations listed where one, or both the resin & filler were 100%-replaced with a bio-derived alternative in a standard commercially produced SMC or BMC formulation as used in the automotive industry. The properties of the standard SMC and BMC grades are included in Table 4 for comparison.

| A Representative Selection of The Formulations Tried | Disposablity (Comparable %Wt Bio Derived) | shrinkage (mm/m) | Density (g/cm$^3$) | Flex Strength (Mpa) | Flex Modulus (GPa) | Impact Resistance (Charpy)(KJ/m$^2$) |
|---|---|---|---|---|---|---|
| **SMC** | | | | | | |
| **Standard SMC** | **0%** | **1.4** | **1.962** | **164** | **10.8** | **60** |
| Resin matrix 100% replaced with Bio-Resin (P55) | 13% | 1.4 | 1.943 | 153 | 10.2 | 54 |
| +Filler replacement with starch flours (e.g. Corn) | 63% | 1.21 | 1.5 | 142 | 7.02 | 81 |
| Standard resin matrix100% filler reinforced with Corn/wood flour | 42% | 1.3 | 1.5 | **166.5** | **9.5** | **91** |
| Standard resin matrix100% filler reinforced with Corn flour | 42% | 1.3 | 1.5 | **197** | **10.3** | **74** |
| Resin, Fibre (Rayon) & Filler (Corn Flour) replacement | 86% | 3+ | 1.28 | 62 | 3.7 | 59 |
| | | | | | | |
| **BMC** | | | | | | |
| **Standard DMC** | **0%** | **1.2 -2.2** | **1.95** | **75** | **12.1** | **17.6** |
| Resin matrix replacement with Bio-Resin (P55) | 9% | ------ | 1.93 | 60 | 11.8 | 16 |
| +Filler replacement with starch flours (Corn) | 66% | ------ | **1.4** | **65** | **6.8** | **35** |
| +Filler replacement with starch flours (Arrowroot) | 66% | ------ | 1.4 | 48 | 5.5 | 13.5 |
| +Filler replacement with starch flours (Tapioca) | 66% | ------ | 1.4 | 61 | 6.1 | 15 |
| +Filler replacement with starch flours (Rice 5um) | 66% | ------ | 1.4 | 49 | 6.5 | 13.5 |
| +Filler replacement with Starch/Wood powder blend | 66% | ------ | 1.4 | **68** | 7.9 | **18.25** |
| Resin, Fibre (Rayon) & Filler (Corn Flour) replacement | 81% | ------ | 1.3 | 45 | 4.5 | 17.6 |

**Table 4** Summary Data for a range of SMC and BMC grades containing Bio-derived ingredients.

Table 4 also details the density values for each formulation and the degree of bio-derived content (expressed as a weight %). Comparing mechanical properties (Flexural modulus, Flexural strength and Charpy impact Resistance), clearly, SMC and BMC grades containing bio-filler demonstrate a mechanical performance that matches or surpasses the standard. Of particular note, is the extremely high impact resistance of starch/wood flour blends – over 50% higher than the standard $CaCO_3$ filled SMC grade. Comparing the results in Table 4 with the somewhat lower performance requirements for end-use parts (Table 1), it is evident that grades containing the Palapreg 55 bio-resin can successfully meet these performance criteria. Bio-filler replacement and combined bio-filler/bio-resin replacement in SMC grades is generally successful in matching or surpassing performance criteria for end-user parts, and can deliver around 25% density reduction, plus bio-derived content of up to 63%. The Bio-derived content was upped further by incorporating Rayon fibre in place of glass. However, the resulting composites fall short in terms of flexural strength and modulus –due to the generally lower stiffness and strength of the these fibres compared to glass. Ultimately, the success of natural fibres in SMC may depend on the selection of the right applications. Researchers at the University of Toronto's Centre for Biocomposites and Biomaterials Processing compared their findings with the mechanical requirements of various SMC auto components. They found that SMC prepared with a combination of 45% hemp fibres and 5% glass fibres offered a tensile strength (over 80 MPa) and modulus (2 GPa) that are the "same or better than" requirements for body parts such as rear lift-gates and front fenders [5]. Similar findings are reported here, where bio-derived grades can be produced to meet mechanical performance criteria for semi-structural parts.

## 6. Conclusions.

SMC is a used widely in very many industry sectors, e.g. construction, marine, kitchen ware, domestic appliances, electrical switching, the power industry, for manholes, tanks, and shaft liners in civil engineering applications, as well as in transport. This diversity of uses, will also involve an accompanying diversity of property requirements, and considering the somewhat lower stiffness/strength performance of natural fibre and rayon grades, it is highly likely that these grades could find application in the wider market.

Key findings reported include the viability of bio-fillers as lightweight alternatives to conventional $CaCO_3$ particulates in SMC and DMC. The importance of controlling paste viscosity has been illustrated and a method for adjusting or tuning the viscosity using filler combinations has been presented. It was been shown that viable SMC/BMC grades can be produced using up to 85% bio-derived ingredients, where the most successful grades demonstrate comparable mechanical performance to standard SMC/BMC, but with a 25% weight saving and 60% bio-derived content.

## 7. Acknowledgements.

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