RECYCLING OF END-OF-LIFE TEXTILE MATERIALS BY FABRICATION OF GREEN COMPOSITES

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ABSTRACT

Material recycling requires rather pure and non-contaminated waste streams, which can be converted by technical processes into new materials and products by economically and technically feasible techniques. It is also required that there is a secondary market for these recycled materials, and preferably in use areas with high economic value. End-of-life textiles cannot yet be recycled as a raw material in a feasible way. Structural composites are an excellent use area for recycled materials, as they are durable and light-weight products, with excellent mechanical properties.

In this study, denim woven fabrics as end-of-life textiles were employed to improve the mechanical and thermal properties of high bio-content epoxy resin. Entropy Resin, manufacturer of commercial bio-based epoxy resin, claimed that bio-based content of system is 37 % and the resin is derived from by-products of industrial processes including wood pulp and biofuel. Bioepoxy was used as bio resins in composite manufacturing, and a conventional polyester resin served as a reference material. To create a wide scope of possibilities the composites were manufacturing using the four techniques: (1) vacuum infusion (VI), (2) resin transfer moulding (RTM), and (3) hand lay-up (HND). To determine the suitability for structural applications the biocomposites were tested for their mechanical and thermal properties. Mechanical tests for tensile, flexural strength and impact behaviour were conducted on composites. Moreover, viscoelastic properties of the composites were evaluated through dynamic mechanical analysis (DMA). Fabricated composites were characterised regarding porosity, water absorption and analysed through microscopic images of the composite cross section. Different manufacturing technique showed varying results. For bioepoxy both HND and VI give superior mechanical properties over RTM, as the latter gives a higher void content, and lower tensile and flexural properties.

1 INTRODUCTION

In this modern day society where fashion trends follow each other rapidly, there is a vast amount of textile waste. This waste consists of textiles that have been purchased and that are disposed of because they are obsolete, textile waste in factories, and textile goods that have not been purchased. For instance 15% of fabric meant for clothing ends up as waste during cutting in the production process. Also 10-20% of textile products are estimated to be wasted each year, where in Hong Kong alone approx. 253 tons are sent to the landfill [1]. Textiles ending up on landfills will be incinerated for energy recovery, meaning that these materials cannot be reprocessed. This is especially alarming if one thinks about i.e. the amount of water needed to produce one denim jeans. The resources at hand need to be dealt with sparingly to raise sustainability and reduce waste. Another problem is that many products nowadays are made from petroleum-based materials, which are harmful to the environment through their production process as well as through their disposal. Also petrochemical resourced are limited as they are derived from fossil fuels, meaning that research into substituting materials is needed [2].

These problem settings are combined in this research, where denim waste is repurposed into biocomposites, which could replace certain petroleum based products, decreasing the need for

petrochemical materials, as well as reducing the amount of denim waste in landfills. Most denim fabric is a woven twill fabric, consisting of 100% cotton; therefore it can be of excellent use in composite manufacturing. Not much research has been done in this area. Wei et al. [3] report composites of discarded denim fabric and polypropylene resin, created by either hand lay-up or hot pressing. These composites have 84 wt% denim. Good mechanical properties are reported with tensile and impact strength of 57 MPa, and 5.1 J/mm, respectively. Lee et al [4] researched the effect of denim added to a poly(lactic acid) matrix in a range of fabric layers from 1-3. Results show that the composite with 3 layers of denim has good mechanical properties with impact, tensile strength and tensile modulus at 82 J/m, 75.75 MPa, and 4.65 GPa, respectively. It is also stated that this composite can substitute traditional composites with a reinforcement of glass fibre, or carbon. Zonatti et al. [5] created composites from 30% recycled denim fibres in combination with an epoxy resin, a polyester resin, and a polyurethane resin. Compared to the neat resin all composites have improved their mechanical properties. The best properties were obtained by the denim/polyester resin, where a 2-fold increase is seen in the tenacity and a 3-fold increase in the modulus, with 13.8 MPa, and 0.772 GPa, respectively.

A bio resin can be from biomass products, microorganisms, biotechnology and petrochemical products. A bioepoxy is a thermoset epoxy resin originating from renewable resources. This bioepoxy is made from renewable plant-based carbon instead of petrochemical carbon. Entropy Resins uses coproducts or waste products of other industrial processes. Entropy Resins [6] explains that their way of producing requires less energy and also produces less harmful by-products. Compared to traditional petrochemical epoxy resins the SuperSAP resin has a biocontent of 37%, and produces 50% less greenhouse gas emission.

The aim of this research is to create biocomposites from bio resins and recycled cotton fabric (denim jeans) through resin transfer moulding, hand lay-up, and vacuum infusion and test their mechanical and thermal properties so that suitability for structural applications can be determined accordingly. To research this aim there are certain questions that need answering.

2 MATERIALS AND METHODS

2.1 Materials

The post-consumer denim jeans were provided by Texaid in Switzerland. The supplied denim consisted of 100% cotton. As denim is widely manufactured of cellulose, and this is a natural source, there is a high variation in properties, which could be problematic for structural applications. Moreover the jeans are second hand, meaning they all have a different level of wear and tear, causing further variation to the properties, but no further preparations were done for the jeans. Two composite samples include a layer of jute to facilitate impregnation. The characteristics of denim and jute fabrics are mentioned in Table 1.

Fabric	**Linear density (per cm)**		**Weave type**	**Weight (g/m^2)**
	Warp	Weft		
Denim	21 (3)	29 (2)	Z twill (1/3)	420 (43.8)
Jute	4	3	Plain	240

Table 1 Denim and Jute fabric characteristics

Bioepoxy has a bio content of 37% (Bio based carbon content, ASTM D6866). It has a mixed viscosity of 0.6 Pa·s, and a measured density of 1.09 g/mL at 25 ˚C. The ratio is which the SuperSAP ONE epoxy and SuperSAP ONE hardener are mixed is 2:1, or 100:47 in weight ratio, and after mixing the resin has a pot life of 25 min at 25 ˚C. (Entropy resins Inc., 2017) The manufacturing of one composite necessitates approximately 100 ml of resin, which then has an epoxy/hardener ratio of 70:33.

A polyester resin and its hardener (Biltema) was used for initial testing to determine the rate and degree of impregnation for RTM. It also serves as a reference in composite analysis. The resin has a

density of 1.12 g/mL at 25 ˚C (SubsTech, 2017). The polyester is combined with 2 wt% of a hardener, and will cure in 30 minutes in room temperature.

2.2 Methods

Sample preparation

As all composite samples are composed of 4 layers of fabric the fibre volume fraction was not controlled precisely. The prepared composites have a fibre weight fraction ranging from 38 to 45 %. The fabric samples were cut from the denim jeans by hand and dried in the oven for at least 24h at 80˚C to ensure moisture evaporation, to lower the possibility of voids in the composites. The sample size for RTM, VI and HND are the same with a diameter of 175 mm. Most composites consist of 4 layers of denim except for two samples (PES-3D1J-RTM and EPOXY-3D1J-RTM) that have 3 layers of denim combined with 1 bottom layer of jute.

Manufacturing techniques

There is a wide array of manufacturing techniques for thermoset biocomposites, comprising of both open and closed mould compositions. Several techniques have been used for sample manufacturing, as the focus is on creating a composite with good mechanical properties and not on the technique with which these are manufactured.

Resin transfer moulding has been used in combination with both polyester and bioepoxy. For the samples with the added jute, the impregnation was done by vacuum, whereas the samples with 4 denim layers were preimpregnated by hand, and placed in the mould. After sample placement and reaching vacuum state the sample was left in the mould until resin gelation, in order for as many air bubbles as possible to be removed by the vacuum. Subsequently both bioepoxy and polyester samples were demoulded and placed in an oven for 10 min at 150 ˚C for post-curing (Table 2).

Only one sample was created with hand lay-up, namely EPOXY-4D-HND. This sample was made as a comparison to RTM and VI, to see whether air bubble removal through vacuum gives divergent results. Naturally these sample layers were pre-impregnated by hand, after which they were stacked one by one, while streaking over the new layer to possibly facilitate air bubble removal. The sample was placed in an oven for 10 minutes at 150 ˚C for post-curing directly after impregnation.

Vacuum infusion moulding was used in the creation of samples of bioepoxy. These samples were used to analyse a difference between RTM and VI for bioepoxy. To ease resin flow a layer of jute was placed on top of the release fabric on the sample stack. The bioepoxy sample was cured upon demoulding. The bioepoxy composites were cured in an oven for 10 minutes at 150˚C for post-curing.

Sample	Resin	Material	Layers	Fibre weight fraction (%)	Fibre volume fraction (%)
PES-3D1J-RTM	Polyester	Denim and jute layer	4	39.9	32.2
PES-4D-RTM	Polyester	Denim	4	41.0	33.3
EPOXY-3D1J-RTM	SuperSAP ONE	Denim and jute layer	4	37.7	29.6
EPOXY-4D-RTM	SuperSAP ONE	Denim	4	39.4	31.3
EPOXY-4D-HND	SuperSAP ONE	Denim	4	38.00	34.5
EPOXY-4D-VAC	SuperSAP ONE SuperSAP ONE	Denim	4	43.7	30.6

Table 2 Composition of samples

Characterization

Test samples were cut by use of a water jet as this causes least damage to the samples. After cutting the samples were dried in an oven at 80˚C for 48h. Then before testing, the specimens were conditioned at 23 ˚C and 50% relative humidity for at least 24 h according to ISO 291 [7].

To determine moisture absorption properties the samples were placed in a conditioner for a month with 100% relative humidity at 60˚C. The void content was measured by the buoyancy method, and microscopic images were taken in order to examine the composite structure, surface and its' void placement.

Tensile specimens of composites were prepared in agreement to the ISO 527 [8]. Tensile tests were performed using a universal H10KT testing machine supplied by Tinius Olsen Ltd., Salfords, UK. Samples were tested at a rate of 10 mm/min.

Three-point flexural tests were conducted according to ISO 14125 [9] using a H10KT testing machine). Flexural tests were performed at a crosshead speed of 10 mm/min and a span length of 64 mm. Five specimens were prepared and analyzed.

In impact testing the charpy energy of a material is measured, which is the amount of energy that is absorbed by the sample before fracture and can be used to determine the material's fracture resistance, which can be related to the material's toughness. Testing was performed according to ISO 179 [10] using a Zwick test instrument.

Dynamic mechanical thermal analysis (DMTA) measures a composite's mechanical viscoelastic properties. The sample was tested in the dual cantilever bending mode for the strain at multi-frequency on a Q series TA instrument. The temperature interval was -20 ˚C to 150 ˚C with a heating rate of 3 ˚C/min, together with a frequency of 1 Hz. Sample dimensions were: length 35 mm, width 10 mm, and thickness 2 - 4 mm depending on the composite type.

Statistical comparisons, based on a one-way analysis of variance (ANOVA) at the 95% confidence level, were performed to test the effects of different fibres on the mechanical properties.

3 RESULTS & DISCUSSION

The effects of resin type, manufacturing technique and addition of jute were investigated by means of tensile, flexural, and impact testing as well as DMTA, moisture absorption and void content.

3.1 VOID CONTENT

When looking at the effect of the addition of jute on the void content it is seen (Figure 1) that for both polyester and bioepoxy jute leads to a lower void content. In polyester the jute even decreases the void content more than 2-fold from 4.3% to 2.1%. In bioepoxy the addition of jute leads to a small decrease in void content from 4.8% to 4.2%. For polyester the void content is slightly lower than bioepoxy.

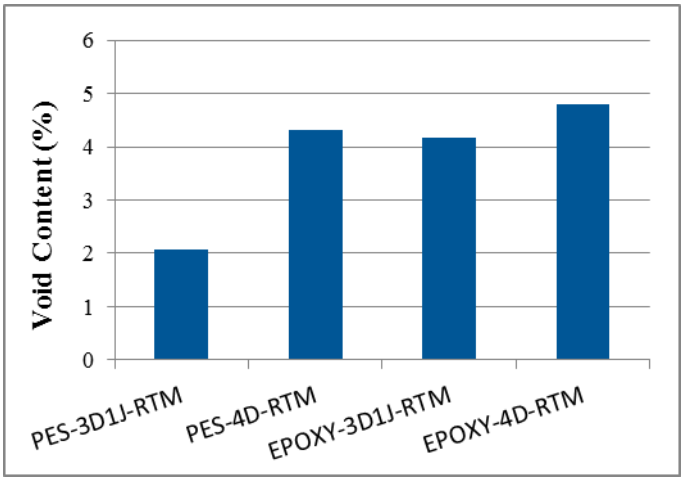

Figure 1: Void content of polyester resin and bioepoxy both with and without the addition of jute

The influence of manufacturing technique for bioepoxy can be seen in Figure 2. RTM (4.8%) clearly has a much higher void content than either HND (1.5%) or VI (0.2%). One-Way ANOVA test showed that void content of materials investigated were significantly different (P-value < 0.05). VI gives the lowest void content, which can be explained by the curing process in vacuum at room temperature, whereas RTM composites were removed from the mould to be cured in the oven.

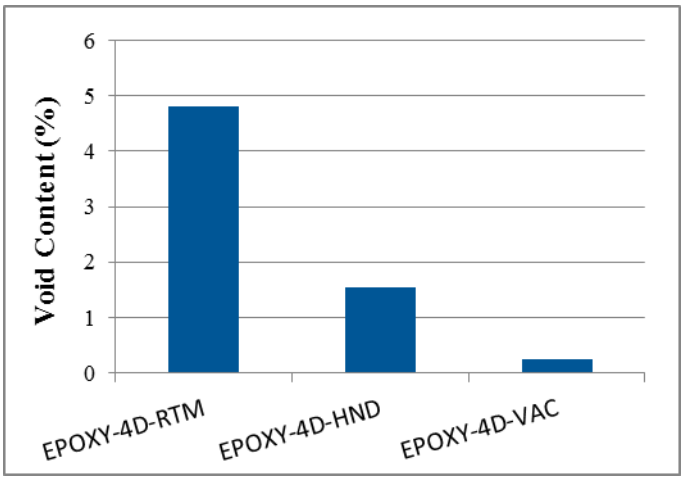

Figure 2: Influence of manufacturing technique on the void content of bioepoxy

3.2 MOISTURE ABSORPTION

The highest moisture absorption percentage (MA%) is seen in polyester with 27% for PES-3D1J-RTM and 22% for PES-4D-RTM. The addition of jute leads to a lower MA% in both polyester and bioepoxy. When comparing manufacturing techniques it is seen that for bioepoxy both RTM and VI with 4 layers of denim have a slightly higher MA%, than RTM with jute, and HND. Again statistical analysis shows there is no significance, meaning that for bioepoxy the manufacturing method does not influence the MA%.

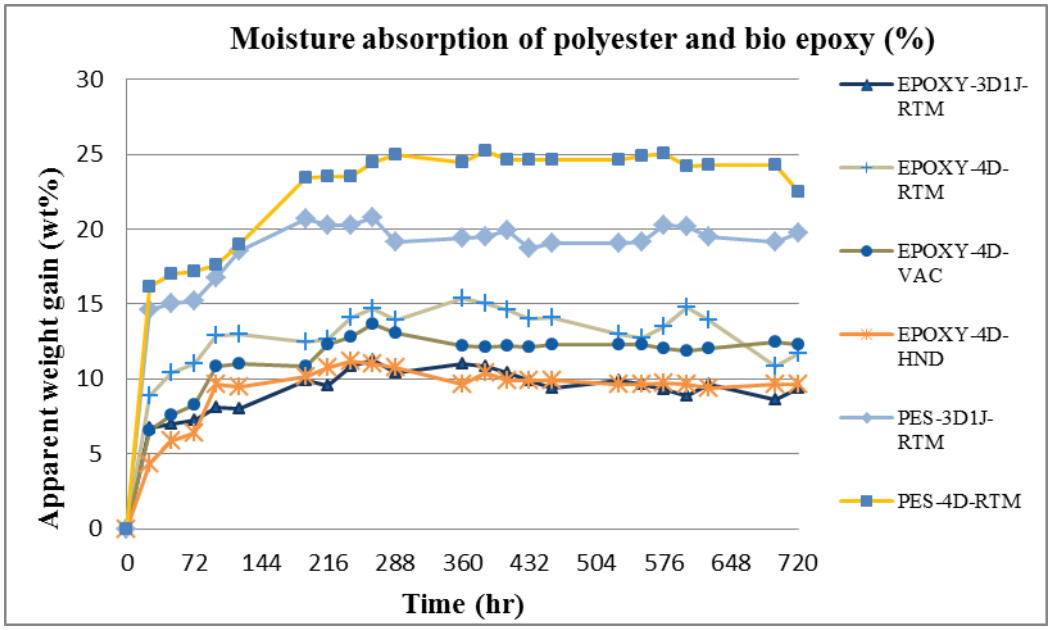

Figure 3: Moisture absorption comparison between PES and bioepoxy composites over a period of 30 days in 100% relative humidity at 60 ˚C

3.3 MECHANICAL PROPERTIES

Tensile testing

When analyzing the effect of the resin type on the tensile properties (Figure 4) it can be seen that bioepoxy gives both the higher modulus and tensile strength. The addition of jute has a negative effect on the polyester with a decrease of 2.2 GPa, while it has a positive effect on the bioepoxy with an increase in modulus by almost 2 GPa. PES-3D1J-RTM is significantly different (p-value < 0.001) from the other samples. For tensile strength statistical analysis measured a significant difference between polyester resin and bioepoxy resin (p-value < 0.001). There is however no significance in the addition of jute to either resin.

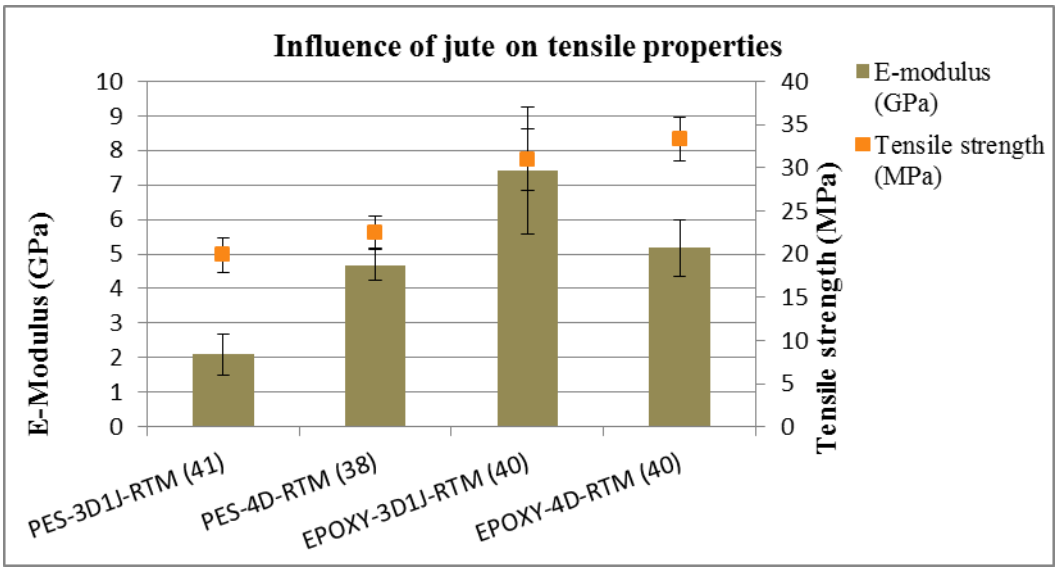

Figure 4 Comparison of the effect of polyester and bioepoxy as well as the addition of a jute layer on the tensile properties

Addition of approximately 40 wt% denim fabric into the bioepoxy resin resulted in a two- to threefold increase in the tensile modulus compared to pure resin, as can be seen in Figure 5. Tensile strength and elongation at max however both decreased significantly, with the exception of the tensile strength of EPOXY-4D-HND. Statistical analysis shows that both Young's modulus and tensile strength for RTM differs significantly from both HND and VAC composites (p-value = 0.001), while HND and VAC do not differ from each other.

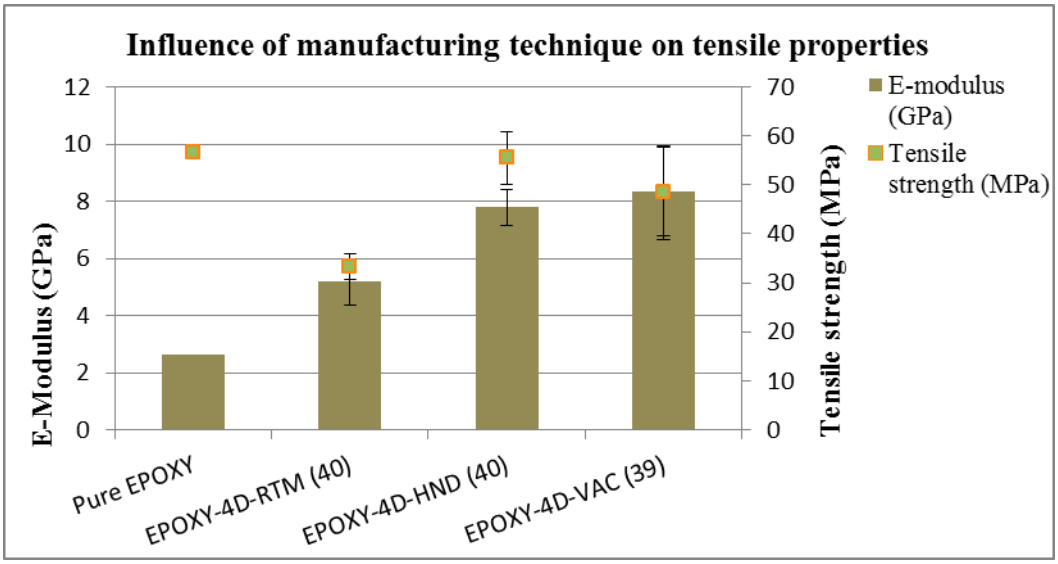

Figure 5 Influence of manufacturing technique on the tensile properties of the bioepoxy resin

Flexural testing

Statistical analysis shows there is no significant difference in modulus for either polyester or bioepoxy with or without jute (p-value > 0.5) (Figure 6). This means that the addition of jute does not influence flexural modulus in any way. For the flexural strength there is a significant difference in addition of jute to bioepoxy (p-value < 0.005), but no significance in addition of jute in polyester. When comparing polyester resin to bioepoxy, Figure 6 shows that bioepoxy has both a higher flexural modulus and strength. Statistical analysis confirms a significance in modulus for EPOXY-4D-RTM, but not for EPOXY-3D1J-RTM (p-value < 0.001). For flexural strength there is a significant difference between polyester and bioepoxy (p-value < 0.001). The lack of significance for bioepoxy with jute comes from a relatively high SD, caused by differences in the composites providing the test samples.

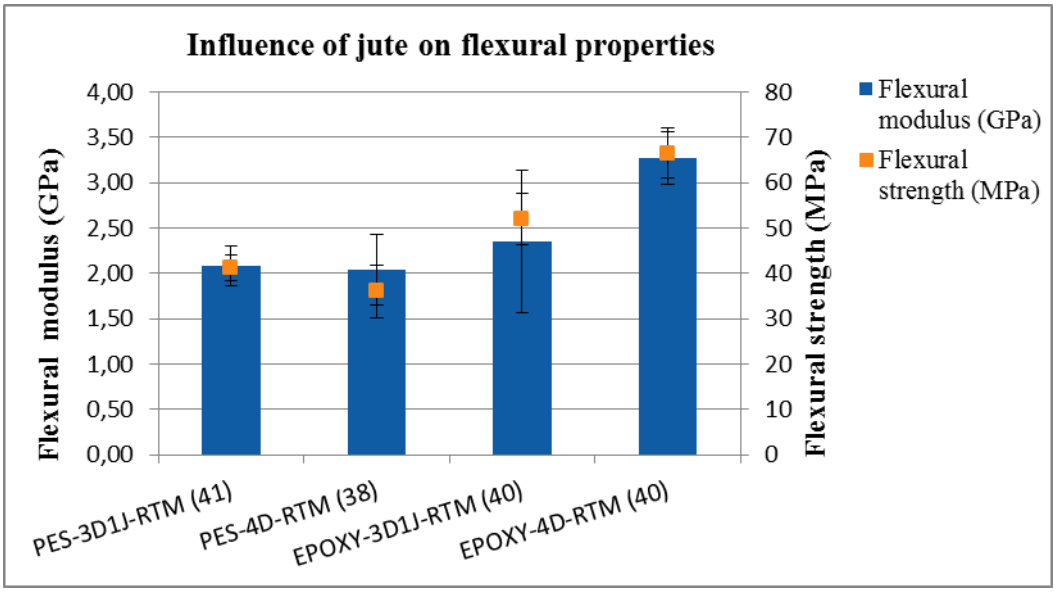

Figure 6 Influence on resin types and the addition of jute on flexural properties

When comparing the bioepoxy composites to the neat resin (Figure 7) it can be said that the flexural modulus is increased overall. For the flexural strength a decrease is seen in RTM composites, whereas both HND and VI give an increase. Statistical analysis indicates no significant difference in modulus between manufacturing methods (p-value > 0.1). For the flexural strength however a significant difference is seen between EPOXY-4D-RTM and EPOXY-4D-HND (p-value < 0.001). This leads to the conclusion that for bioepoxy there is no difference in manufacturing techniques for the flexural modulus, but there is for the flexural strength.

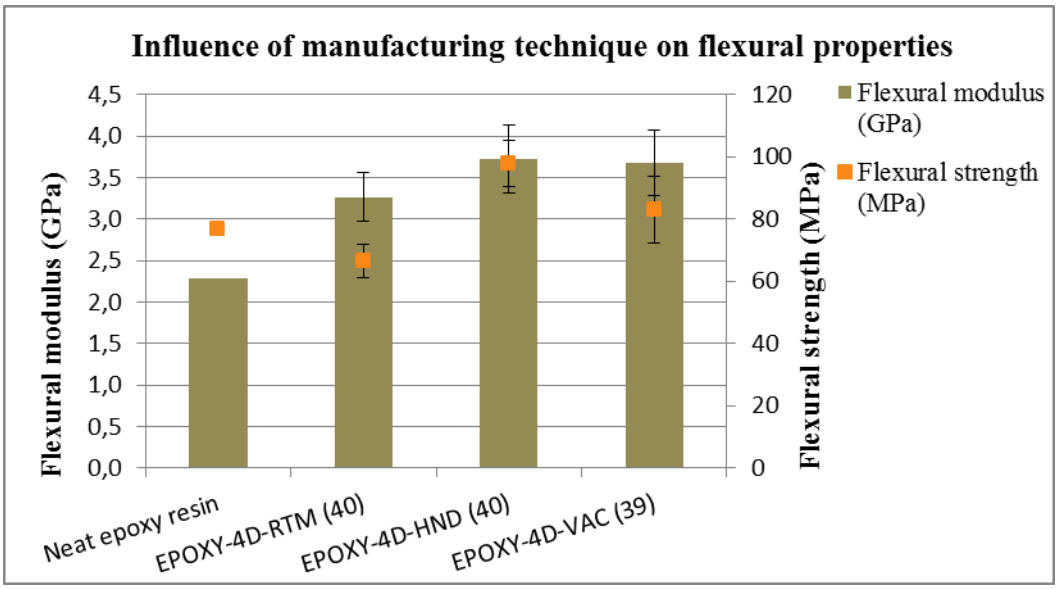

Figure 7 Influence of manufacturing technique in bioepoxy on flexural properties

Impact testing

Addition of jute has a negative effect on impact properties as can be seen in Figure 8. When comparing polyester to bioepoxy it is seen that polyester has a higher impact resistance. This is also in line with tensile and flexural testing as these showed a higher flexibility for

unsaturated polyester composites. Statistical analysis shows significant difference for the addition of jute to bioepoxy (p-value < 0.001), but no significance for the addition of jute to polyester. This results in the addition of jute not making a difference for the polyester resin, where it does decrease the impact resistance of bioepoxy.

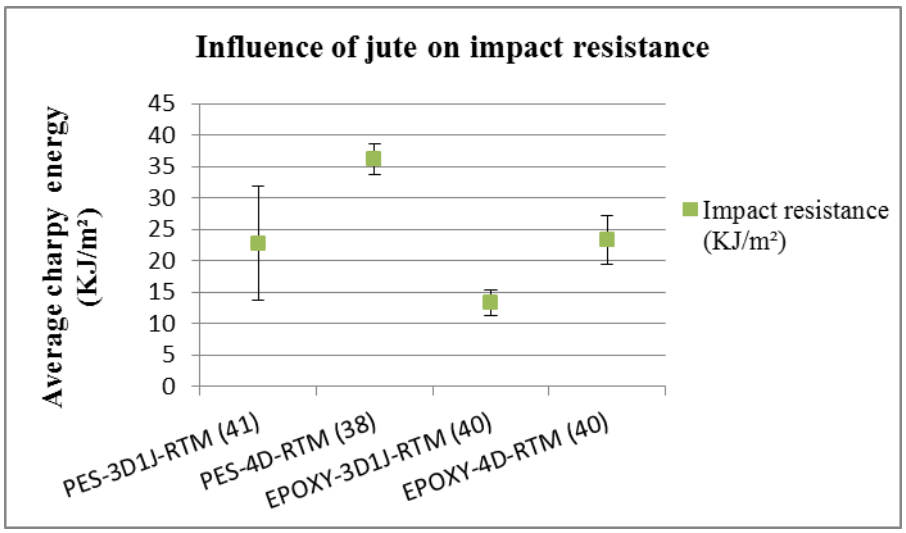

Figure 8 Influence of resin type and addition of jute in both polyester and bioepoxy on impact resistance

As can be seen from Figure 9, hand lay-up gives the lowest impact resistance, which can be accredited to a relatively high void content as there was no vacuum present during composite production. The highest impact resistance is from RTM. Statistical analysis shows a significant difference between HND and RTM (p-value = 0.003), but no significance for VI. This means that HND gives bioepoxy composites the lowest impact resistance.

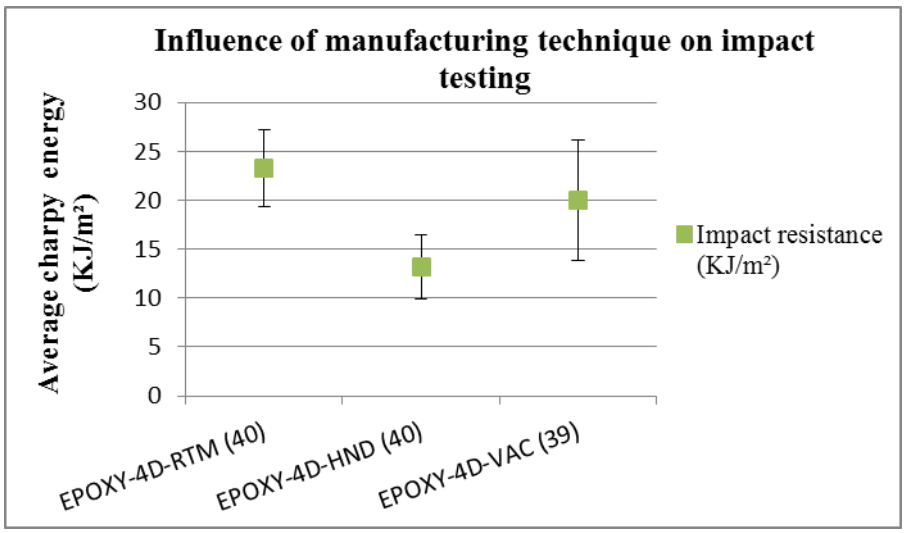

Figure 9 Influence of the manufacturing technique of bioepoxy on impact resistance

3.4 THERMAL PROPERTIES

Dynamic mechanical thermal analysis

As can be seen in Figure 10, the addition of jute seems to increase the E' of polyester, while it significantly decreases the E' of bioepoxy. When comparing the tan δ it is seen that polyester has a higher T_g than bioepoxy, as well as a slightly wider peak.

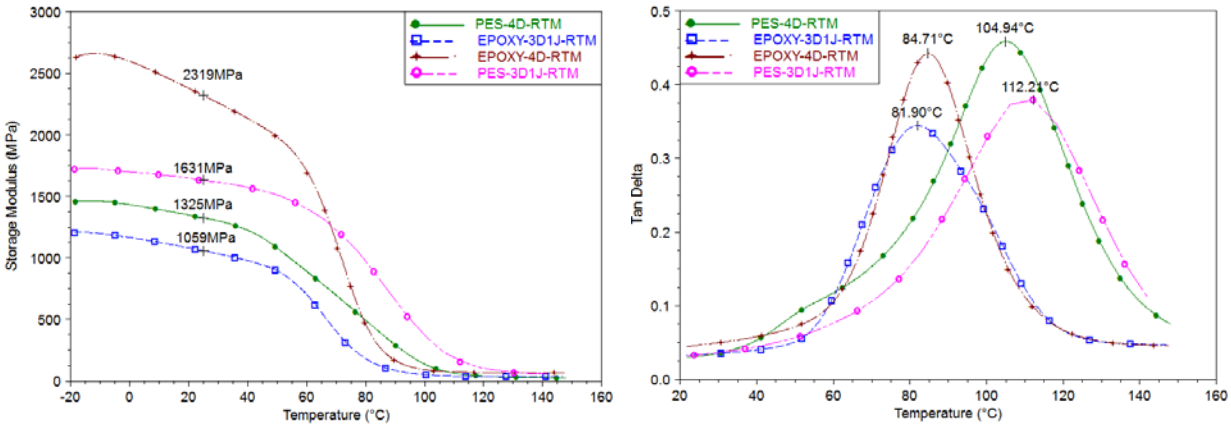

Figure 10 Influence of addition of jute and resin type on the E' and tan delta

3.5 Microscopic images

Figure 11 A and B shows that bioepoxy without jute has many interlaminar voids, whereas the one with jute does not. The interlaminar voids and lack thereof can be explained by the difference in impregnation methods. EPOXY-3D1J-RTM was impregnated by RTM, while EPOXY-4D-RTM was impregnated by hand and placed in the RTM mould for void removal. EPOXY-3D1J-RTM has an irregular surface due to the structure of the jute. This can also explain the high void content measured by the buoyancy method. It is also seen that dry spots are located inside the yarn, meaning this either did not have enough time or enough resin to impregnate completely.

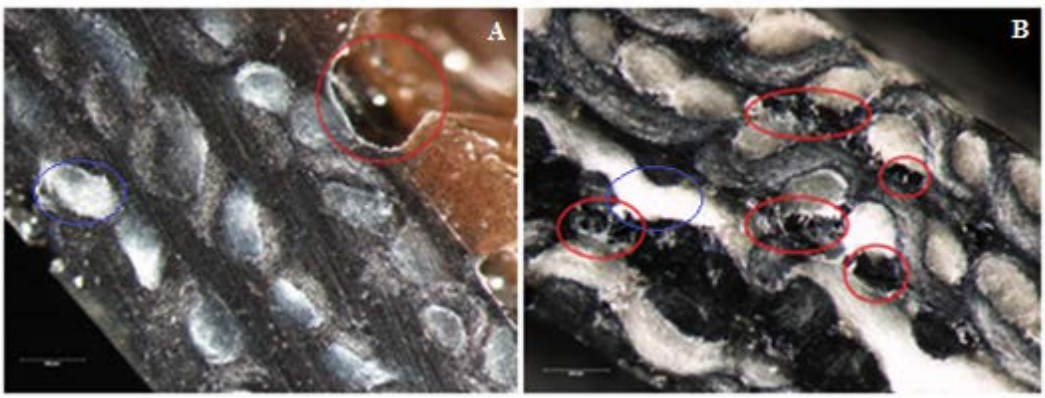

Figure 11 Microscopic comparison (3x) of bioepoxy with jute (A) and without jute (B), manufactured by RTM

As both bioepoxy composites in Figure 12 were impregnated by hand, the effect of RTM on void removal can be seen. Buoyancy measures a void content of 1.54% and 4.8% for EPOXY-4D-HND, and EPOXY-4D-RTM, respectively. EPOXY-4D-HND has a high void content, and the interlaminar voids present have a considerable size. There are also many small dry spots spread out over the composite layers. Both of these can be explained by the short impregnation time, as the composite was

cured in the oven right after impregnation. EPOXY-4D-RTM also shows interlaminar voids, but of much smaller size, due to both the void removal by RTM and a longer impregnation time.

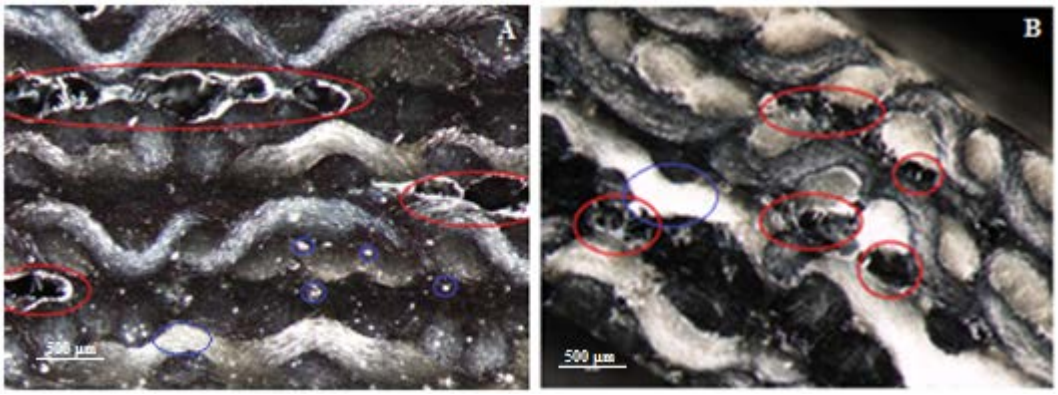

Figure 12 Microscopic comparison (3x) of EPOXY-4D-HND (A) and EPOXY-4D-RTM (B)

4 CONCLUSIONS

With the fashion industry being the way it is, a vast amount of textile waste is created each year. In order for this to pollute the earth as little as possible other solutions need to be found, than letting it end up in a landfill. A fair amount of research is going into recycling with the purpose of the material being re-used in new textiles, but the technology is not yet sufficient to ensure materials with a good quality. Mostly the materials are down-cycled into a secondary market with lower quality demands and economical value. Also these recycling plants cannot yet handle the large waste stream. In this study the possibility has been researched into repurposing denim waste into green composites for structural applications, which in turn are suitable for a secondary market.

There are several bio resins that would be suitable for green composite manufacturing, as this is a highly researched area. The chemical composition of these resins has been compared, and a bioepoxy resin has been chosen for analysis, as literature showed great promise. Most previous research into denim waste for composite manufacturing resulted in biocomposites with a petroleum based matrix. This research takes composite manufacturing one step further into green composites composed of recycled material and a bio resin.

The curing of the resins proved to be troublesome with both the polyester resin and bioepoxy curing before full impregnation. For bioepoxy and polyester this resulted into a somewhat higher void content and moisture absorption as the resin had insufficient time to impregnate completely into the center of the yarn.

As stated before, this study aimed to create green composites for structural applications. Test results of the manufactured composites have been compared to literature in order to determine suitability. Analysis resulted in composites from different manufacturing methods implementable in structural applications such as automotive interior parts, furniture, interior construction and sports, and leisure equipment. These composites are EPOXY-3D1J-RTM, EPOXY-4D-HND and EPOXY-4D-VAC.

As this research has shown denim waste can be reused into green composites for a secondary market with a high economical value. A benefit of being able to use the textiles itself instead of fibres is that there are relatively low investment costs for manufacturing. This results in a higher amount of textile waste being reused instead of ending up on landfills or being incinerated. Also lots of water is used in the denim consumer lifecycle. This lifecycle can now be extended significantly, meaning the H_2O usage is spread out over a longer period, decreasing denim's water footprint. As denim is a classic material in fashion, a constant stream of denim waste is expected, which will not suddenly stop.

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