Towards reinforcement solutions for urban fibre/fabric waste using bio-based biodegradable resins.

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Abstract. The main research question is how to systematically define and characterize urban textile waste and how to effectively utilise it to produce reinforcement(s) with selected bio-based biodegradable resin(s). Several composite samples have been produced utilising predominantly natural and predominantly synthetic fibres by combining loose fibres with PLA, nonwoven fabric with PLA, woven fabric with PLA, two-layer composite & four-layer composite samples. Physico-chemical characterisations according to the established standards have been conducted. The present work is a step toward the circular economy and closing the loop in textile value chain.

1. Introduction
Close loop thinking is an important part of current business strategies to reduce resource consumption and positively contribute to the environment [1]. Consuming resources and generating waste on large scale is typical for mainstream manufacturing industries [2]. ‘Circular economy’ (CE) is an approach that would transform the use of resources in the economy and waste would become a valuable input to other processes and products. Textile industry has huge impact on the environment. Around 90Kton textile waste is collected in the Netherlands. Around 35% thereof is damaged and not re-usable anymore as such [3]. Huge amounts of urban waste could serve as raw material sources for the new applications. However, urban waste is less defined and consists of multiple materials. Therefore, the research question in this paper is: how to systematically define and characterize urban textile waste and how to utilise them effectively to produce reinforcement(s) with selected biodegradable bio-based resin(s).

2. Materials and Methods

2.1 Physicochemical Characterisation
Characterisation of the textile fibres/fabric have been conducted to establish the fibre type and percentage of each type in the waste stream. Physicochemical characterisation according to the established standards have been conducted in terms of Raw materials test (AATCC TM 20A-2014), Microscopic images (AATCC TM20-2013), Burn test (AATCC TM20-2013), Moisture regain (AATCC TM 20A-2014), Oil extraction to determine the percentage of oil in the fibres (Mesdan Lab, Code 273B), DuPont Waterdrop test (AATCC TM79-2014) density, fibre/fabric thickness, weight per
m² (ASTM D3776-07), Elongation and tensile strength (ISO 2062:2009). The primary purpose of these characterisation/tests were to identify % of fibres in textile waste streams and suitability of substrates for composite making. Additionally, several synthetic waste streams have been tested with FT-IR (Bruker-Tensor 27) for polymer determination.

2.2 Nonwoven production
In order to understand the effect of dimensional stability of reinforcement on composite making. Some of the loose waste fibres have been converted into non-woven using air-laid web forming and needle punching technique to ensure almost equal strength in all direction. Nonwoven production was carried out at Havivank BV, Tilburg, the Netherlands.

2.3 Composite production
Hot press technique (preheating at 200°C for 40min and pressing with 2MPa for 3-5 min) has been used for composite samples using three different poly lactic acid resins (PLA 2003D granulates which is high molecular weight and crystalline, PLA 4060D granulates which is low molecular weight and amorphous and PLA nonwoven with unknown properties) from Rodenburg Biopolymers BV and Havivank BV, The Netherlands. Resulting composite samples have been evaluated using Charpy test (ISO 14125:1998) for toughness/energy uptake and Three-point bending test (ISO 179-1:2010) for the flexural strength.

3. Results and Discussion

3.1 Characterisation of textile waste streams
A systematic approach has been adopted (Figure 1) to handle the textile waste streams. Physiochemical tests and characterisation (see section 2.1) have been conducted to identify the % of certain fibre type in the textile waste stream. As there are hardly no pure waste streams, the textile waste has been broadly categorised as A) Predominantly natural fibres, further divided in to three parts a) recycled fibres; such as Jute, Cacao Jute, Denim, Cotton, Acrylic wool blends, workwear mix fibres, b) dust; such as cotton dust, Acrylic wool dust, and c) woven fabrics; such as jute bags and Denim jeans. For the B) Predominantly synthetic fibres samples are e.g. waste stream 1 (claimed as Polyamide 6/6.6), waste stream 2 (claimed as Polypropylene fibres), waste stream 3 (claimed as Polyethylene) and waste stream 4 (shredded fibres from workwear/uniforms) etc. Considering the complex and heterogeneous mix from batch to batch, the fibre/textiles waste streams have been used as such, since it is almost impossible to purify them into 100% natural fibres and 100% synthetic fibres.

Figure 1: A systematic approach to handle fibre/textile waste into composite samples.

Physiochemical characterisation of several textile waste streams (dust, fibres, non-woven, woven) samples have been conducted according to the tests and standards mentioned in the section 2.1. However, it was still not possible to establish the fibre types from the synthetic waste streams. Therefore FT-IR studies of three different synthetic waste streams (WS1, WS2, WS3) have been conducted using similar reference materials. The results of FT-IR spectra are presented in Figure 3.
Figure 2: FT-IR spectra of A) reference Polyamide vs WS1-waste stream 1, B) reference Polypropylene vs WS2 – waste stream 2, and C) reference Polyethylene vs WS3-waste stream 3.
It is clear from the Figure 3A that important absorption peaks of amide (3292 cm\(^{-1}\) and 1631 cm\(^{-1}\)) are not seen, when looking at the waste material of PA 6/66 (WS1). The remarkable peaks in the waste material (WS1) are at 1713 cm\(^{-1}\) and 1235 cm\(^{-1}\). These peaks together are characteristics of an ester group. From comparing the spectra of the waste material, it is clearly not a polyamide. The spectra of WS1 has more similarities with PES, therefore it can be said that the main component of WS1 is polyester and not as proposed polyamide spectra.

Figure 3B shows the comparison FT-IR spectra of reference Polypropylene and waste stream 2. The spectra of the PP reference show three important peaks around 2916 cm\(^{-1}\), 1453 cm\(^{-1}\) and 1375 cm\(^{-1}\). The peaks at 1375 cm\(^{-1}\) and 1453 cm\(^{-1}\), are similar to the absorption frequency of CH\(_2\) bending, these two peaks form an alkane group. It can be said that the main component is a PP of the reference material. It is clear that there are additional peaks at 1713 cm\(^{-1}\) and 1235 cm\(^{-1}\) resembling to polyester (Figure 3B). Hence it can be said that the waste stream 2 is a mixture of PP and PES.

Figure 3C, shows the FT-IR spectra of PE starting with the reference and then waste material (WS3). The spectra of the reference PE show three important peaks, at around 2912 cm\(^{-1}\), 2845 cm\(^{-1}\) and 1461 cm\(^{-1}\) the peaks at 2912 cm\(^{-1}\) and 2845 cm\(^{-1}\) are similar to the absorption frequency of CH stretch with a frequency around 2850-3000 cm\(^{-1}\). The peaks of the PE waste material are similar to the reference PE. With peaks around 2913 cm\(^{-1}\), 2847 cm\(^{-1}\) and 1471 cm\(^{-1}\). From spectra of the waste material compared to the reference material it can be assumed that the main component is PE of the reference and of the waste.

Finally, woven jute bags, loose cacao jute fibres, and cotton mix have been selected for the further study from predominantly natural fibre waste stream. For the synthetic fibres waste stream the chosen materials are waste stream 1 (PES), Waste stream 2 (Polypropylene/PES mix) and Uniforms/Workwear mix. The decision was made on the basis of availability of raw materials and ease/possibility to make in composite,

3.2 Predominantly natural fibre based composite production

On the basis of availability of raw materials and possibility to make composite, three different samples have been selected for the further study. The samples are woven jute bags, loose cacao jute fibres, and cotton mix. Composite samples are produced with two-layer and four-layer reinforcement. The idea is to check following two hypotheses, i) if converting loose fibres into non-woven delivers composite with higher strength, ii) if doubling the amount of reinforcement layers leads to double the mechanical strength. The selected results are presented in Figure 3.

| Composite | 1. Cacao jute/cotton non-woven with PLA web | 2. Cacao jute/cotton non-woven with PLA web | 3. Woven jute bag with PLA web | 4. Prepressed loose denim fibres with PLA web |
|-----------|------------------------------------------|------------------------------------------|---------------------------------|------------------------------------------|
|           | Two layers                               | Four layers                              | Four layers (Highest bending properties) | Four layers (Highest energy uptake) |

| Bending modulus (N/mm\(^2\)) | 2673 | 3228 | 5680 | 750 |
| Charpy test strength (in J) | 0.39 | 0.78 | 0.37 | 1.68 |

**Figure 3:** Predominantly natural (loose, non-woven and woven) reinforcement samples.
As described earlier, the Three-point bending test (ISO 179-1:2010) is meant for flexural strength and the Charpy test for the toughness/energy uptake. As seen from the figure 3, it is clear that doubling the reinforcement layers (sample 1 vs sample 2), not necessarily doubles the bending modulus, however the energy uptake tends to increase linearly. The higher flexural strength/bending properties (5680 N/mm²) achieved by woven jute bag and PLA as resin. The woven structure of the reinforcement seems to be responsible for this results. The highest toughness/energy uptake (in J) is achieved for prepressed loose denim fibres. This can be attributed to easier penetration of PLA resin across the loose denim fibers during composite production. To sum up it is advisable to use loose fibre reinforcement for rigid surface like tables. Woven reinforced materials such as the jute bag could be used for chairs as their bending behavior is superior. Additionally, it is to say that mixing Jute and Cotton in nonwoven structure (Figure 3, sample 2) could be a good solution for making a product with mixed/moderate properties.

### 3.3 Predominantly synthetic fibre based composite production

Several composite samples have been produced utilising predominantly synthetic fibres by combining loose fibres with PLA, nonwoven fabric with PLA. Figure 4 shows selected composite samples made out of predominantly synthetic loose fibres vs. non-woven. The hypothesis was that additional dimensional stability of reinforcement would provide more mechanical strength. Results showed that converting the loose fibres into non-woven doesn’t necessarily increase the mechanical strength. This is attributed to the fact that, the melted PLA resin can easily penetrate around loose fibres compare to non-woven reinforcement. The choice of PLA resin certainly showed influence in terms of mechanical strength. Composite (sample C1 = 1.45 J) made with PLA 2003D granules showed higher mechanical strength, attributed to its high molecular weight and crystallinity compared with PLA 4060D (0.25 J) and PLA web (0.31 J). Additionally, samples produced with nonwovens reinforcement gave highest flexibility to polypropylene composite. (2919 N/mm²).

| Loose fibre reinforcement | Nonwoven reinforcement |
|--------------------------|------------------------|
| **A.** WS1-Polyester (loose) | **A1.** WS1 – Polyester (nonwoven) |
| [0.08 J, 635 N/mm²] | [0.11 J, 1050 N/mm²] |
| **B.** WS2-Polypropylene (loose) | **B1.** WS2-Polypropylene (nonwoven) |
| [0.19 J, 2327 N/mm²] | [0.18 J, 2919 N/mm²] |
| **C.** WS4-Work wear mixture (loose) | **C1.** Work wear mixture (nonwoven) |
| [0.38 J, 2512 N/mm²] | [1.45 J, 2713 N/mm²] |

**Figure 4:** Predominantly synthetic reinforcement samples (loose vs non-woven).
4. Conclusions
During the course of this project, we identified a great number of possible material combinations with varying technical and mechanical characteristics, as well as with a wide range of different perceptual values (all data not shared). With some of the most favourable material combinations first prototypes are developed, tested and investigated. Several loose fibre/non-woven/woven reinforced composite samples have been prepared using three different PLA resins as a matrix for bio-based composites. Composite samples with several interesting combinations with workwear mix fibres, Polypropylene non-woven, Jute fibres, Denim fabric etc., have been produced with diverse properties (strength and flexibility). It is possible to get tailored made end properties in terms of strength, flexibility by selecting right type of reinforcement material and choice of resin. However further systematic research is ongoing in the direction of optimizing end properties and circularity of the composite product(s). There are opportunities in the area of i) large number of end applications, ii) the higher added value on the material properties, iii) large volumes (with use of textile based waste materials) can be achieved with and iv) the ability to optimize for (full) circularity. The present work is a step toward the circular economy and closing the loop in textile value chain.

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