FIBRE LENGTH DISTRIBUTION OF SHREDDED THERMOPLASTIC COMPOSITE SCRAP

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ABSTRACT

Shredding is a crucial step when recycling thermoplastic composite waste. The outcome of this step can be in the form of flakes or particles of various size, which strongly depend on the chosen shredding solution and the material type. It was shown that the mechanical properties of a part manufactured with these flakes are influenced by the fibre length of those. Characterising the length of fibres in the flakes is therefore important to link it with mechanical properties [1]–[4]. Literature on characterising FLD in the case of large (a few centimetres) and multi-layered flakes is scarce. However, recycling solutions for these flake sizes exist [5], confirming the interest for this topic. In order to fill this gap, this paper firstly develops a method to determine the FLD of a batch of flakes and secondly investigates the effect of both process parameters and waste size on the FLD. The newly developed method is based on the image processing of flakes. Besides, the sampling process that is linked to the image processing method was found to be both repeatable and reproducible at a high precision, showing that batch-process recycling is robust with respect to the influence of sampling. Following that, ways to tailor the FLD were explored. It seems that the scrap size does not influence the FLD but blade width and screen size of the shredding machine largely govern this FLD.

1 INTRODUCTION

The thermoplastic composite (TPC) market has been growing in the past decades and is expected to continue to do so in the near future. One direct consequence is the increase in production scrap and end-of-life waste. As a result, the development of recycling solutions is incited for economic, environmental and legislative reasons [6]. All the recycling solutions for fibre reinforced plastics follow a route with similar steps: (1) collection of the waste at production sites; (2) size reduction of the waste to particles; (3) sorting or separation of the impurities, which may also be done before size reduction or at the collection; (4) manufacturing of a component, with processes that depend on the chosen solution [7]. It was shown that the size reduction technology does not only depend on the type of waste but also on the required input for re-manufacturing such as sorting easiness or particle size [8]–[11]. Regarding the latter, literature shows interest for various recyclate: powder used as fillers [10], chopped TPC semipreg [12]–[14], short fibres [13] and shredded TPC laminates [5]. In all cases, the recycled product is a discontinuous fibre reinforced composite. The retained fibre length is a parameter that partially governs mechanical properties of the recycled product [15], [16] and flow behaviour during its processing [12], [17]. It is therefore crucial to have fibre length characteristics of the comminution outcome in order to further study the rheology of such material or its mechanical properties.

Various methods are currently used to characterise fibre length distribution (FLD) in discontinuous fibre composites but they suffer from drawbacks. Many methods are time-consuming, destructive

techniques or only work on a selection of materials [4], [18]. In particular, literature is scarce with regard to measuring FLD of large chopped or shredded flakes such as in [5], [12].

This article proposes a different method to measure the FLD of large (from here on: a few centimetres) shredded laminate waste. A first section will review the literature on this topic and current alternatives. This new method will then be described, analysed and used to measure the FLD of shredded TPC laminate waste. The results will enable the determination of the influence of shredding parameters and waste size on the FLD. Hence, guidelines to the shredding process will be given to tailor the FLD to a particular need.

2 LITERATURE REVIEW

Several methods were developed to analyse the FLD of either recycled or discontinuous fibre composites. Turner [4] and Palmer [19] classified shredded fibrous materials with commonly used methods for granular materials: multiple stage sieves or air classifier. Shredded fibrous materials were separated and collected at each sieving stage or classifying step. Regarding sieving, particles falling through a sieve having a hole size of α but staying above the following sieve β have an average size ranging from α to β . FLD was considered to be similar to the particle size distribution (PSD) in [4]. A problem shown by Turner is that the slenderness of the particles, which can be high in the case of prepreg or fabric, can bias the results. Elongated fibrous flakes can fall through fine sieves even though their fibre length is longer than the sieve size. Palmer et al. [19], who used air classifiers, considered the PSD by characterising it using automatic image processing. In this case, most particles were elongated fibre bundles thus fibre length was similar to particle length. However, this cannot be extended to flakes that have a fabric structure and are multi-layered.

Methods for short and long fibre composites comprise fibre reclamation and image processing of spread fibres, which are captured using a scanner for instance [1], [18]. Even though these methods give accurate results, they are time consuming because every fibre in the selected composite area is characterised. This can add up to the treatment of several thousand fibres. Hence, only a small area of the specimen can be analysed and it requires that the specimen has a FLD evenly distributed over the specimen.

Another method is based on 3D tomography of a selected specimen [20]. It gives the length, the dispersion, distribution and orientation of fibres within the specimen. However, the specimen size is limited by the spatial resolution and the number of pixels in the sensor. The voxel size must be smaller than the fibre diameter to accurately describe every single fibre. Additionally, measuring and computing times are longer than other methods thus a limited amount of specimens can be treated.

When considering shredded TPC scrap, it seems there is an interest for the recycling of large flakes as shown in [5], [21]. This recycling solution produces long fibres and variable flake sizes when industrial size reduction systems are used. Therefore, solutions such as 3D tomography and analysis of reclaimed fibres are suitable to little extent. Additionally, the analysis of flake sizes using multiple sieves may give inaccurate results as explained by Turner. As a consequence, an alternative method is required. A method is proposed in Section 4 of this article for further analysis of shredded material.

Category	Average weight [g]		
C/PPS offcuts - small	8		
C/PPS offcuts - medium	164		
C/PPS offcuts - large	328		
C/PPS offcuts - trims	n.a.		

Table 1: classification of the various offcuts used in this study.

3 MATERIALS AND SHREDDING TECHNOLOGY

One type of scrap material was collected for this study: quasi-isotropic carbon/PPS consolidated offcuts from the trimming stage of a stamp-forming process (hereinafter referred to as C/PPS offcuts), as shown in Figure 1 (a). Noteworthy, the C/PPS offcuts had various size and thickness, and were classified in three categories based on their unit weight as shown in Table 1. This table also shows other types of offcuts was used in this study.

The size reduction technology chosen to process the offcuts was multiple-shaft shredding. Several shredders from Untha were used: S20, RS30, RS40 and RS50. These systems have a low rotational speed (20 to 30 rpm) [22], [23], which prevents the production of fine particles thanks to the limited energy of impacts of particle on machine and particle on particle [24]. Hence, almost only the desired flake size is produced. An example is given in Figure 1 in which C/PPS offcuts are shredded with an S20: most flakes seem to have the same size and no fines are present. Figure 2 illustrates the comminution mechanisms of four-shaft shredders and shows an inside view of such machine. When incoming material falls in the shredder, several teeth covering the blades grab it in the blue areas (hatched regions at the entry – illustrated in Figure 2). The material is thus forced to go downwards and is sheared between the overlapping blades (orange areas – hatched regions on overlapping blades). In order to ensure that the outcome is composed of the desired PSD, a screen is usually placed right below the blades. If the flakes are too large to go through, they move upwards, thanks to the two rotating outer shafts, and are shredded one more time. Thus, both the blade width and the screen size seem to have an influence on the PSD. Other parameters such as clearance between blades, sharpness of the teeth or small variations of rotational speed were shown to have an influence on the fracture mechanism but little on the PSD [24]. Therefore, the present article will only focus on the blade width and the screen size, as shown in Table 2. Several other parameters vary between the four shredders used in this study such as the number of shafts (4 shafts for RS machines, 2 for the S20), shaft diameters, the maximum torque and the throughput. The last three only affect the size and volume of the input material but not the PSD. Similarly, the two outer shafts in four-shaft shredders enable auto cleaning of the shredding chamber but does not influence the PSD.



Figure 1: Pictures of C/PPS offcuts (a) and the outcome after shredding (b).

Shredder	S20	RS30	RS40	RS50
Blade width [mm]	19	19	19	29
Screen size [mm]	-	-, 40	-, 40	-, 25, 40

Table 2: Shredders used in this study and their influencing parameters. The various screen sizes listed indicate that multiple screens were used for different tests.

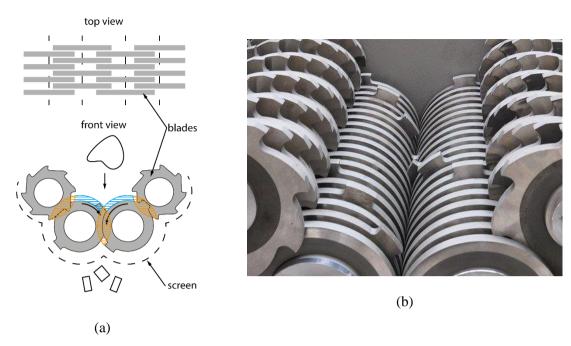


Figure 2: (a) Schematics of a four-shaft shredder from the top and front, (b) inside view of a four-shaft shredder (image courtesy of Untha GmbH).

4 METHODS

A new method is proposed to analyse the FLD of shredded TPC waste such as C/PPS offcuts. According to Section 2, it requires to be an accurate and a quick method that works for large consolidated flakes.

Two image processing methods were developed to estimate the FLD of a batch of flakes: a manual method that accurately and precisely produces FLD but has a drawback of taking much time, and an automatic method that is quick but induces small errors. The later differs only on the fibre orientations within flakes. The quasi-isotropic layup of the flakes was arbitrary approximated with a discrete isotropic layup of 18 plies for every 10°. Precautions are to be taken for other flake layups. The various steps for both methods are as follows and are summarised in Figure 3:

Manual

- 1. Sample a batch of flakes from its parent population
- 2. Capture pictures of the flakes using a diffusor box
- 3. Manually indicate the fibre orientation of the top layer if it is not visible for an automatic algorithm (in the software)
- 4. Convert pictures to binary images
- 5. Generate binary arrays of lines that are oriented as the fibre orientations in each flake (all layers are considered)
- 6. Intersect the binary images and their related binary arrays
- 7. Generate the line length distribution of the intersected images

Automatic

- 1. Sample a batch of flakes from its parent population
- 2. Capture pictures of the flakes
- 3. Convert pictures to binary arrays
- 4. Generate binary arrays of lines for a set of orientations θ ($\forall k \in [1,18], \theta = 2\pi k/18$)
- 5. Intersect the binary images and the binary arrays
- 6. Generate the line length distribution of the intersected images

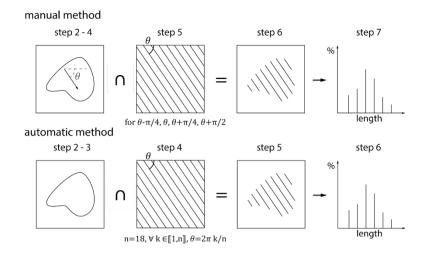


Figure 3: Process steps for both the manual (top) and automatic (bottom) analysing method. Step numbering refers to the descriptive list in Section 4.

Test number	Waste type	Blade width [mm]	Screen size [mm]	Shredder	Remarks
#1	C/PPS trims	19	40	RS30	5+2 batches
#2	C/PPS large	29	-	RS50	No screen, one cut
#3	C/PPS large	29	40	RS50	
#4	C/PPS large	29	25	RS50	
#5	C/PPS large	19	40	RS40	
#6	C/PPS large	19	-	S20	No screen, 5 cuts
#7	C/PPS medium	19	-	S20	No screen, 5 cuts
#8	C/PPS small	19	-	S20	No screen, 5 cuts

Table 3: List of the various performed shredding tests.

5 RESULTS AND DISCUSSION

C/PPS offcuts were shredded with the machine settings listed in Table 3. First, the FLD of C/PPS offcuts were analysed with both the methods to determine the statistical variability of the automatic method explained in Section 4, as well as for the sampling process. Second, the automatic method was used to analyse shredded material for which shredding parameters and offcut size are varied.

5.1 Method accuracy

In both methods defined in Section 4, sampling the batches was random and may induce errors in the calculated FLD compared to the true FLD of the population. Repeatability and reproducibility studies were performed on the shredded waste. A total of 7 batches were taken off from the same population of flakes (C/PPS offcuts - trims): 5 sampled by a first operator on one day (numbered 1 to 5), 2 sampled by a second operator on another day (numbered 6 and 7). The population of flakes weighted approximately 5 kg and was stored stationary until sampling, to prevent any granular convection. Additionally, all batches were sampled without replacement.

Both the manual and automatic methods were used to calculate the FLD in these batches. Examples of the FLD calculated with the manual and automatic methods are shown in Figure 4. It is noted that the three displayed distributions are almost equal. A correlation coefficient, based on the Pearson correlation coefficient, was use to quantify their correlation:

$$r_{x,m} = \frac{\sum_{i=1}^{n} (x_i - \tilde{x})(y_i - \tilde{y})}{\sqrt{\sum_{i=1}^{n} (x_i - \tilde{x})^2 \sum_{i=1}^{n} (y_i - \tilde{y})^2}}$$

Where x refers to the x^{th} batch, $r_{x,m}$ is the correlation coefficient for the manual method, x_i is the volume fraction of the batch x for the fibre length i, \tilde{x} is the arithmetic mean of all x_i and y refers to all seven batches combined. Each batch is then compared to all seven batches combined. Table 4 summarises the correlation coefficients for all seven batches, as well the number of flakes per batch. It shows that all batches are extremely close to the mean, with $r_{x,m}$ varying from 0.968 to 0.997, even for a small number of flakes per batch (from 61 to 93). Several conclusions can already be drawn from this table:

- The sampling method is shown to have low statistical variability.
- The sampling method is both repeatable (batches 1 to 5) and reproducible (batches 6 and 7). Therefore, a single batch seems enough to have precise results.
- While looking at the full recycling loop, if the flakes are converted to a new component with a batch-process, such as in [5], it is important to have a similar FLD from batch to batch. Here, the batches of flakes, even when containing a small number of flakes, have similar FLD. This can improve the robustness of such a recycling solution.

Following that, the automatic method was compared to the manual method by a similar coefficient of correlation:

$$r_{x,a} = \frac{\sum_{i=1}^{n} (x_{i,a} - \widetilde{x_a})(x_{i,m} - \widetilde{x_m})}{\sqrt{\sum_{i=1}^{n} (x_{i,a} - \widetilde{x_a})^2 \sum_{i=1}^{n} (x_{i,m} - \widetilde{x_m})^2}}$$

Where the x_a variables correspond to the FLD calculated with the automatic method for the x^{th} batch and the x_m variables correspond to the FLD calculated with the manual method for the same x^{th} batch. Table 4 shows that the correlation between the automatic and manual FLD is very strong for each batch. The error added by using the automatic method is therefore extremely limited.

As a result, the following part of the study will be only performed with one batch per type, which will be analysed with the automatic method only.

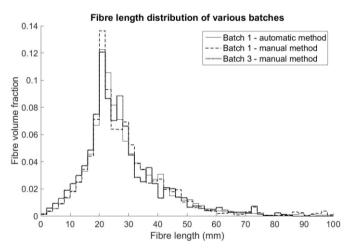


Figure 4: Comparison of FLD of various batches. The automatic method gives a FLD close to the manual method for batch 1. Batches 1 and 3 are part of the repeatability study, in which several batches are sampled from the same flake population. Their FLDs are close to each other.

Batch	#1	#2	#3	#4	#5	#6	#7
$r_{x,m}$	0.992	0.996	0.968	0.996	0.997	0.991	0.986
Flakes per batch	63	63	64	61	93	59	60
$r_{x,a}$	0.986	0.990	0.975	0.989	0.961	0.973	0.982

Table 4: Characteristics of the batches used for the repeatability and reproducibility study.

5.2 Influence of shredding parameters

C/PPS offcuts of three various sizes defined in Table 1 were shredded with an S20 machine. The machine does not have a screen, which is described in Figure 2, thus the outcome was manually added back in the S20 hopper to simulate the effect of having a screen. The final shredded outcome was passed five times through the shredder. One batch was taken for each of the three sizes (small, medium, large) and analysed. The calculated FLDs are shown in the first three rows of Figure 5 in a boxplot form with a highlight for the peak location. Firstly, it shows that the peaks are all located at 20 mm, which is the approximate blade width (19 mm). Secondly, the boxplots slightly shift to longer fibre length when the offcut size increases. However, this is very limited compared to the difference between the offcuts sizes which vary from 8 g to 328 g. Thirdly, the spread of these three FLD does not vary, which implies that the maximum fibre length is similar for all three offcut sizes.

Next to these three tests, several others were carried out with RS30, 40 and 50 machines. Large C/PPS offcuts were shredded for the following combination of blade width [mm] and screen size [mm]: (29-25), (19-40), (29-60),

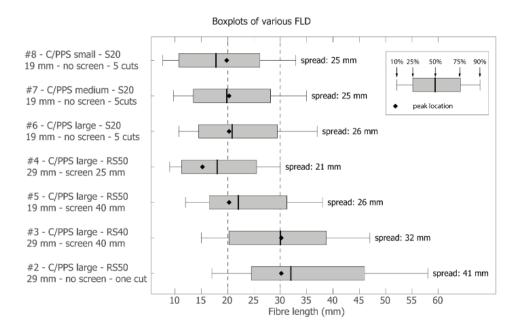


Figure 5: FLD in a boxplot form for various shredded offcuts and for several shredding parameters. Process parameters influence the location and shape of FLD whereas the offcut size barely has an influence.

6 CONCLUSION

The initial aim of this study was to analyse the influence of shredding parameters and offcut sizes on the FLD of shredded consolidated TPC waste. A new method was required to analyse the FLD of a sample of flakes, because current methods do not seem to be able to work with consolidated flakes of a few centimetres. Reliability aspects of this method were first studied: the introduction of errors with the sampling and by the method itself.

Two methods based on image processing were implemented. A manual method, which precisely and accurately calculates the FLD of a set of flakes; and a quicker automatic method, which estimated the

FLD quite accurately with only very limited errors. It was found that the sampling was repeatable and reproducible, as well as precise. Most interestingly, small batches of flakes taken from one population have nearly the same FLD. This property is valuable for batch-wise recycling solutions. It can then be assumed that the FLD of all batches are equal.

Additionally, there is the balance in term of FLD between processibility and mechanical properties for a given recycling solution and a type of material. Shorter fibres usually improves processibility whereas long fibres give higher mechanical properties. It is therefore crucial to be able to transform offcuts to any range of FLD before proceeding to further processing steps. Results of this study showed that the offcut size has a limited effect on the FLD. However, the blade width and screen size of multiple shaft shredders were found to influence both the spread and the mean of the FLD. Hence, it seems that one can work with almost any scrap size indifferently, since the desired FLD is mostly governed by blade width and screen size.

Follow up work will consider these various FLD and determine how they influence a recycling solution developed in [5].

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REFERENCES

- [1] A. Bernasconi, D. Rossin, and C. Armanni, "Analysis of the effect of mechanical recycling upon tensile strength of a short glass fibre reinforced polyamide 6,6," *Eng. Fract. Mech.*, vol. 74, no. 4, pp. 627–641, 2007.
- [2] F. W. J. Van Hattum and C. A. Bernardo, "A model to predict the strength of short fiber composites," *Polym. Compos.*, vol. 20, no. 4, pp. 524–533, 1999.
- [3] F. W. J. Van Hattum, J. P. Nunes, and C. A. Bernardo, "A theoretical and experimental study of new towpreg-based long fibre thermoplastic composites," *Compos. Part A Appl. Sci. Manuf.*, vol. 36, no. 1, pp. 25–32, 2005.
- [4] T. A. Turner, S. J. Pickering, and N. A. Warrior, "Development of recycled carbon fibre moulding compounds Preparation of waste composites," *Compos. Part B Eng.*, vol. 42, no. 3, pp. 517–525, 2011.
- [5] T. A. De Bruijn, G. A. Vincent, and F. W. J. Van Hattum, "Recycling of long fibre thermoplastic composites by low shear mixing," in *SAMPE Europe*, 2016, pp. 1–8.
- [6] European Parliament, "Directive 1999 31 EC on landfill of waste," 1999.
- [7] S. J. Pickering, "Recycling technologies for thermoset composite materials-current status," *Compos. Part A Appl. Sci. Manuf.*, vol. 37, no. 8, pp. 1206–1215, 2006.
- [8] M. Roux, N. Eguemann, C. Dransfeld, F. Thiebaud, and D. Perreux, "Thermoplastic carbon fibre-reinforced polymer recycling with electrodynamical fragmentation: From cradle to cradle," *J. Thermoplast. Compos. Mater.*, 2015.
- [9] J. Palmer, O. R. Ghita, L. Savage, and K. E. Evans, "Successful closed-loop recycling of thermoset composites," *Compos. Part A Appl. Sci. Manuf.*, vol. 40, no. 4, pp. 490–498, 2009.
- [10] T. Inoh, T. Yokoi, and K.-I. Sekiyama, "SMC Recycling Technology," *J. Thermoplast. Compos. Mater.*, vol. 7, 1994.
- [11] R. E. Allred and J. M. Gosau, "Size-reduction considerations for large-scale recycling of composite manufacturing scrap," in *CAMX 2015*, 2015, pp. 1767–1780.
- [12] M. I. Abdul Rasheed, "Compression molding of chopped woven thermoplastic composite flakes: A study on processing and performance," University of Twente, 2016.
- [13] Toho Tenax, "Green Innovations." [Online]. Available: http://www.tohotenax.com/products/green-innovations/. [Accessed: 12-May-2017].
- [14] CarboNXT, "carboNxt chopped." [Online]. Available: http://www.carbonxt.de/index.php?id=166. [Accessed: 12-May-2017].
- [15] M. Selezneva, "Experimental and Theoretical Investigations of Mechanical Properties of

- Randomly-Oriented Strand (ROS) Composites," McGill University, 2015.
- [16] J. L. Thomason, "The influence of fibre length and concentration on the properties of glass fibre reinforced polypropylene: 6 The properties of injectino moulded ong fibre PP at high fibre content," *Compos. Part A Appl. Sci. Manuf.*, vol. 36, no. 1, pp. 995–1003, 2005.
- [17] C. Servais, A. Luciani, and J.-A. E. Månson, "Fiber–fiber interaction in concentrated suspensions: Dispersed fiber bundles," *J. Rheol.* (*N. Y. N. Y*)., vol. 43, no. 4, 1999.
- [18] S. Y. Fu, Y.-W. Mai, E. C.-Y. Ching, and R. K. Li, "Correction of the measurement of fiber length of short fiber reinforced thermoplastics," *Compos. Part A Appl. Sci. Manuf.*, vol. 33, no. 11, pp. 1549–1555, 2002.
- [19] J. Palmer, "Mechanical recycling of automotive composites for use as reinforcement in thermoset composites," University of Exeter, 2009.
- [20] D. Salaberger, K. A. Kannappan, J. Kastner, J. Reussner, and T. Auinger, "Evaluation of computed tomography data from fibre reinforced polymers to determine fibre length distribution," *Int. Polym. Process.*, vol. 26, no. 3, 2011.
- [21] T. A. De Bruijn, G. Vincent, and F. W. J. Van Hattum, "Recycling C/PPS laminates into long fibre thermoplastic composites by low shear mixing," in 21st International Conference in Composite Materials, 2017.
- [22] Untha, "RS30/40 datasheet." [Online]. Available: http://www.untha.com/files/pdf/brochures/en/machines/holz_en_rs30-40-wood-shredder.pdf. [Accessed: 11-May-2017].
- [23] Untha, "RS50/60/100 datasheet." [Online]. Available: http://www.untha.com/files/pdf/brochures/en/machines/rs50-100_industrial-shredder en 8seitig.pdf. [Accessed: 11-May-2017].
- [24] D. Woldt, "Zerkleinerung nicht-spröder Stoffe in Rotorscheren," TU Bergakademie Freiberg, 2003.