EVALUATION AND ENHANCEMENT OF RECYCLABILITY FOR COATED PACKAGING PAPERS

Pieter Samyn[1], Patrick Cosemans[1], Bram Bamps[2], Roos Peeters[2]

[1]Materials and Packaging Research & Services, Imo-Imomec, Hasselt University, Belgium
[2]SIRRIS – Department Circular Economy, Belgium

Original scientific paper
doi: 10.5281/zenodo.10007049
pieter.samyn@sirris.be

Abstract: *In order to meet the European target for recyclability of packaging materials in general, and paper packaging in particular, harmonized guidelines have recently been published to test and assess the recyclability of coated papers, based on repulping and screening of recycled fibers in combination with visual evaluation of the quality of papersheets. Based on a design-for-recycling strategy, a first estimate can be made for the recyclability of current paper coatings, but the constant new developments in the sector and the commercialization of new coating materials require a standardized framework for experimental evaluation of recyclability. In this study, the testing protocol was validated in relation to variations in the repulping conditions such as repulping temperature, and number of revolutions for a selection of industrial packaging papers, including acrylic dispersion coatings or LDPE laminated paper. The testing procedure seems robust as influences of repulping temperature are inferior in the range of 30 to 50°C, while the influence of the number of revolutions is more critical and levels off within a given operation window. The statistical differences between repulpability of non-recyclable and recyclable packaging papers are detected in a benchmarking study against newspaper or tissue paper. For difficult-to-recycling papers, opportunities for improvement of coating removal or valorization of mixed fiber/coating brokes collected in the reject fraction should be further explored. As such, the functionality and circularity of coated papers can simultaneously be enhanced.*

Keywords: packaging paper, coating, recyclability, testing

1 INTRODUCTION

The paper and paperboard materials have become a widespread origin for alternative packaging materials and are more often considered as potential replacement for traditional single-use plastic packaging. They benefit from the renewable origin of cellulose fibres, their high specific strength, flexibility, and light weight. Depending on the needs for food packaging purposes, the paper properties can be adapted towards required barrier properties against water, oxygen, moisture, oils, and aroma by application of a coating (Tyagi et al., 2021). In view of their use in a more sustainable and circular economy, the recycling of coated packaging papers and recovery of the fiber fraction is preferred above biodegradation or composting (Sridach et al., 2006). Indeed, ambitious targets are set both at European and Belgian level. European legislation dictates that 65% of all packaging waste needs to be recycled by 2025 and even up to 70% by 2030 (European Commission, 2020). On Belgian level, the Belgian Food Industry states that all food packaging should be recyclable, reusable, or biodegradable by 2025 (FEVIA, 2021). A renewed focus on paper-based packaging materials has therefore been put in parallel with the development of novel and barrier coatings for better preservation and shelflife of the packed food, while being compatible with the paper recycling process.

The conventional barrier coatings of extruded polyethylene are difficult to recycle as a significant fraction of the paper fibers remains embedded in the PE film and is not easily recovered by repulping (Bilek et al., 2021). Alternatively, the separation of a PLA melt coating may also be problematic due to its strongly hydrophilic properties (Kunam et al., 2022). As a result, both the coating fraction and the recovered fibers are highly contaminated and have low value for recycling, while additional layers eventually need to be developed as an intermediate release coating (Al-Gharrawi et al., 2021). However, the design of multilayer coating systems should be avoided to reduce the complexibility of recycling (Koppolu et al., 2019). A series of waterborne dispersion coatings are promising for recycling, where the emulsion particles will either stick to the fibers or fillers and/or can be more easily separated during the stock preparation via screening (Kathuria et al., 2022). The repulping yield of acrylic emulsion coated papers was reported to be potentially above 99% with little contamination of the wet-end process (Lee et al., 2020). Depending on the composition of the polyacrylate-based polymer, however, the problems with disintegration of the coating on a pilot pulper were reported as a fraction of soft flakes could not be removed by slot screening due to the large deformability of the coating fragments (Lee et al., 2017): consequently, additional mechanical treatment of the accept fraction was needed for fragmentation and removal of the contaminants up to 95%. Alternatively, the bio-based dispersion coatings of hydroxypropylated starch and hydroxypropyl cellulose may also cause high number of rejects (> 50%) being worse compared to synthetic polymer coatings (Ovaska et al., 2017).

Repulping and recyclability testing has a long history and, highly depending on a number of parameters and conditions, it only has recently been harmonized in a European guideline that is currently valid for mills with standard recycling technology (CEPI, 2022). In this study, we have validated the recyclability testing procedures for a series of commercially available coated packaging papers, in order to determine significance, repeatability and sensitivity of the method to operational conditions. Present evaluations provide us with a reference framework for further testing and assessment of paper recyclability.

2 MATERIAL AND METHODS

2. 1 Samples

Four types of industrial packaging paper samples have been introduced as examplatory cases, including a reference uncoated paper (A), 2 recyclable coated paper samples (B, C) and 1 non-recyclable coated paper sample (D). The Kraft paper 60 g/m^2 (A) was used as a reference substrate with a standard 5 g/m^2 proprietary precoating. The two recyclable coated papers contain on a proprietary acrylic dispersion coating with two different compositions and were applied by curtain coating: the sample B has a coating grammage of 10 g/m^2, the sample C has coating grammage of 7 g/m^2. Both the coating thickness and composition for the recyclable samples is different, but further details are not known and not relevant for present validation study. The non-recyclable sample D contains an extrusion coated LDPE film with coat weight of 15 g/m^2 (common folding carton weight).

Furthermore, a benchmarking study was performed for the recycling of a tissue paper grade (sample E, Tork Dry Multi-Purpose Wipes) and uncoated newsprint paper (sample F).

2. 2 Recyclability testing

Assessment of recyclability is done according to the document (version October 2022): “Harmonised European laboratory test method to produce parameters enabling the assessment of the recyclability of paper and board products in standard paper and board recycling mills” (CEPI, 2022). A practical implementation of the testing protocol is schematized in Figure 1.

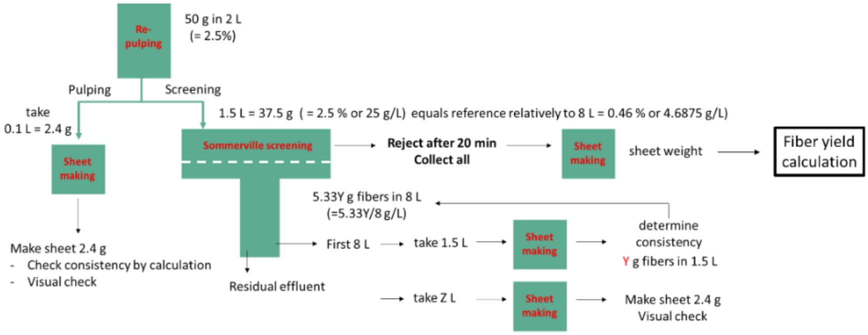

Figure 1. Schematic interpretation of the testing protocol for paper recyclability.

- The repulping is done in a disintegrator compliant with ISO 5263-1 (Figure 2a) on an oven-dry sample of 50 ± 1 g. The 25 x 25 mm^2 cut pieces are diluted with tap water at mildly alkaline pH = 7 to 8. The total volume of sample and water is approximately 2000 g, resulting in a stock consistency of 2.5 %. No pre-wetting or soaking was done. The disintegration time was varied, while a fixed time of 10 min (30,000 revolutions) is requested in the guidelines. The desintegration was done at different temperatures of 52, 40 and 30°C to evaluate the influence on testing results, while an intermediate temperature of 40 ± 1°C is required by the guidelines.
- The screening is done on a Sommerville screen with plates of 5 mm diameter holes (coarse screening) and 150 µm wide slots (fine screening) (Figure 2b), using a pulp sample size of 1.5 liter. The rejects are collected on top of the screen after 20 min to determine their oven-dry weight. The fiber yield is calculated from the oven-dry weight of collected rejects relatvely to the 1.5 liter sample corresponding to 37.5 g virgin pulp.
- The accept and reject fractions from the screening tests are further used for sheetmaking (Figure 2c). Optical evaluation is done under a microscope with lens magnification 5x and stitching of 4x4 fields of view.

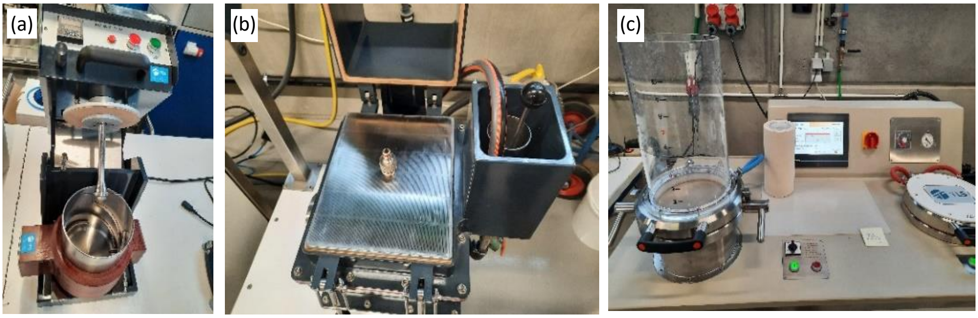

Figure 2. Set-up of pilot line at Sirris (Leuven, Belgium) for paper recyclability testing, with (a) pulp disintegration, (b) screening, (c) sheetmaking in watercolumn and dryer.

3 RESULTS AND DISCUSSION

3. 1 Desintegration – repulping

The influence of disintegration variables on fiber yield for coated paper samples A to D is presented in Figure 3, indicating the effect of disintegration time (Figure 3a) and temperature (Figure 3b). The reference uncoated paper (sample A) has clearly best repulpability with fiber yield of > 99.5 % and little influences of the repulping parameters, only the very low times of 10000 rmp are not sufficient to disintegrate the entire sample. Both dispersion coated papers (samples B, C) present very similar results once a sufficient repulping time of 30000 rpm is applied. As expected, the repulpability for LDPE laminated paper (sample D) is worse with fiber yield of 70 to 85 %. The fiber yield for unrecyclable papers more strongly depends on the repulping conditions as an in-homogeneous recycled pulp is obtained. The effect of temperature change did not affect repulpability in a significant way for recyclable samples B, C. Therefore, it can be concluded that differences in fiber yield are insignificant small and both types of dispersion coated paper can be repulped according to this method with representative fiber yield.

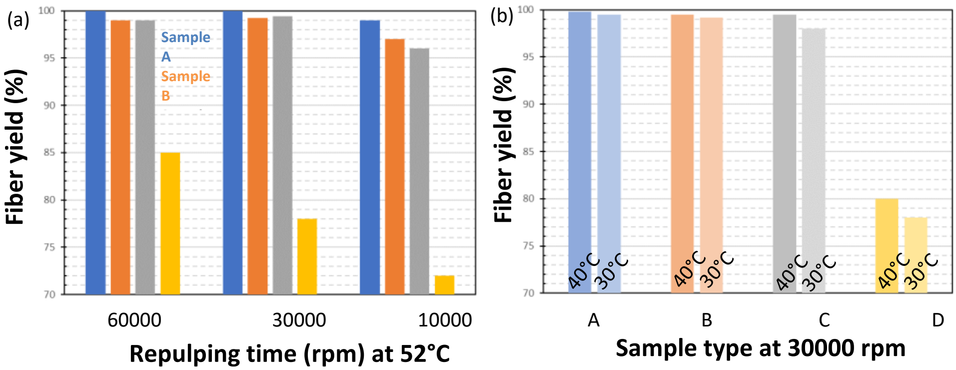

Figure 3. Influence of repulping conditions on fiber yield after fine screening, (a) different repulping times at 52°C, (b) different temperatures at 30000 rpm.

3. 2 Screening

The variations in fiber yield after coarse and fine screening for a non-recyclable coated packaging paper (Figure 4a) and recyclable coated packaging paper (Figure 4b) were determined on five independent sample runs for sample D and B, indicating better reproducibility for recyclable papers (fiber yield min 95.3 %, max 97.6 %, standard deviation 2.3 %) compared to non-recyclable papers (fiber yield min 25.5 %, max 41.0 %, standard deviation 15.5 %), owing to the large influence on screening efficiency by the accumulation of non-recyclable fractions as visualized by a photograph of the rejects on the fine screen. The coarse rejects for non-recyclable papers include broken plastic remainants of the LDPE coating not passing through the grid. In addition, the fine rejects include agglomerated paper fibers with coating substances. The almost full fiber recovery of uncoated paper substrates suggest that wet-end additives and eventual presence of fillers/pigments (proprietary information) did not strongly hinder recyclability in present case.

The results of the benchmarking study for tissue and newsprint paper grades, show that the papers clearly can be categorized with significant statistical differences in the recovered fiber yield (Figure 5). However, the recyclability of tissue paper is inferior dueg to the use of strong wet-end chemicals in the fabrication of tissue paper to provide higher mechanical strength properties. The latter hamper the recyclability process mainly during fine screening, which is different compared to the non-recyclable packaging paper grades that mainly fail in recyclability during the coarse screening process. The fiber agglomerates formed in presence of the strong wet-end chemicals indeed seems to be mainly separated during a fine screening test (sample E).

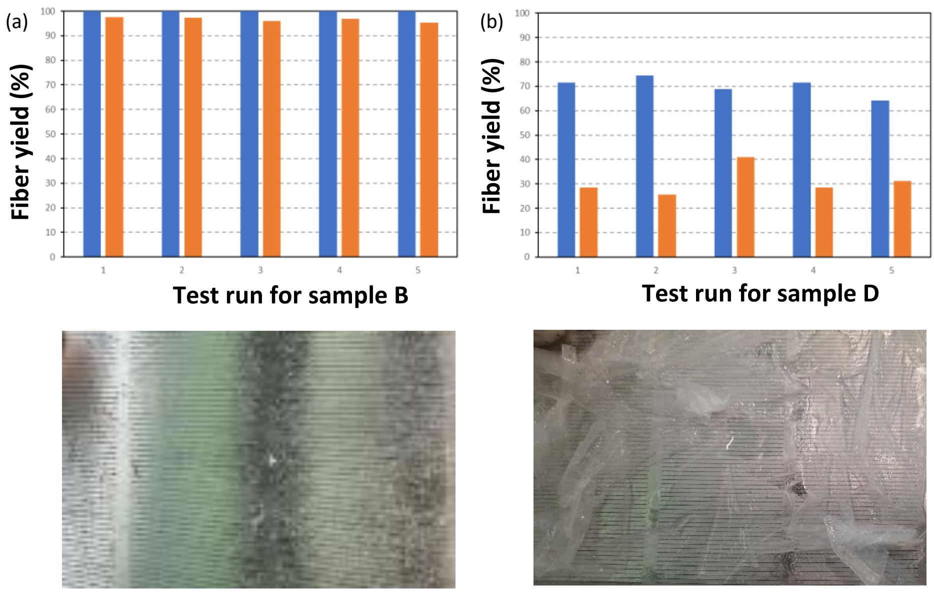

Figure 4. Influence of screening conditions (coarse screening = blue bars, fine screen-ing = orange bars) on fiber yield during repetitive testing and photographs of fine screening rejects for (a) sample B, (b) sample D.

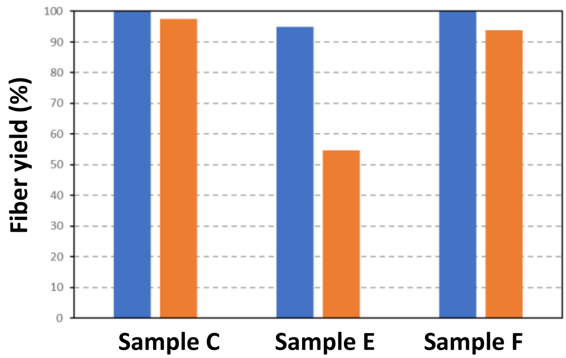

Figure 5. Benchmarking study for fiber yield after coarse screening (blue bars), fine screening (orange bars) of tissue paper (sample E) and newsprint paper (sample F).

3. 3 Sheet evaluation

The handsheets from accept and reject fractions after fine screening of coated packaging papers are evaluated by optical microscopy, providing an overview micrograph (stitching area 4x4) (Figure 6) and detailed views (Figure 7).

For recyclable paper grades (sample B), the sheets from accepts are very homogeneous with still a broad distribution of fiber sizes present comparable to the virgin pulp. Still some particles of impurities originating from broken fragments of the acrylic coating remain present in the accepts, with sizes below 0.5 mm that do not hinder the homogeneous sheetmaking. The large fiber agglomerates observed in the original sample after disintegration are removed and transferred into the reject fraction that forms a weak sheet.

For non-recyclable paper grades (sample D), fibers after disintegration are strongly compacted and transferred into the reject fraction to a large amount, where they are strongly clogged together within a polymer fraction originating from the coating. Also the accepts from fine screening still contain significant amount of residual polymer fractions that were not yet fully removed by the screening process.

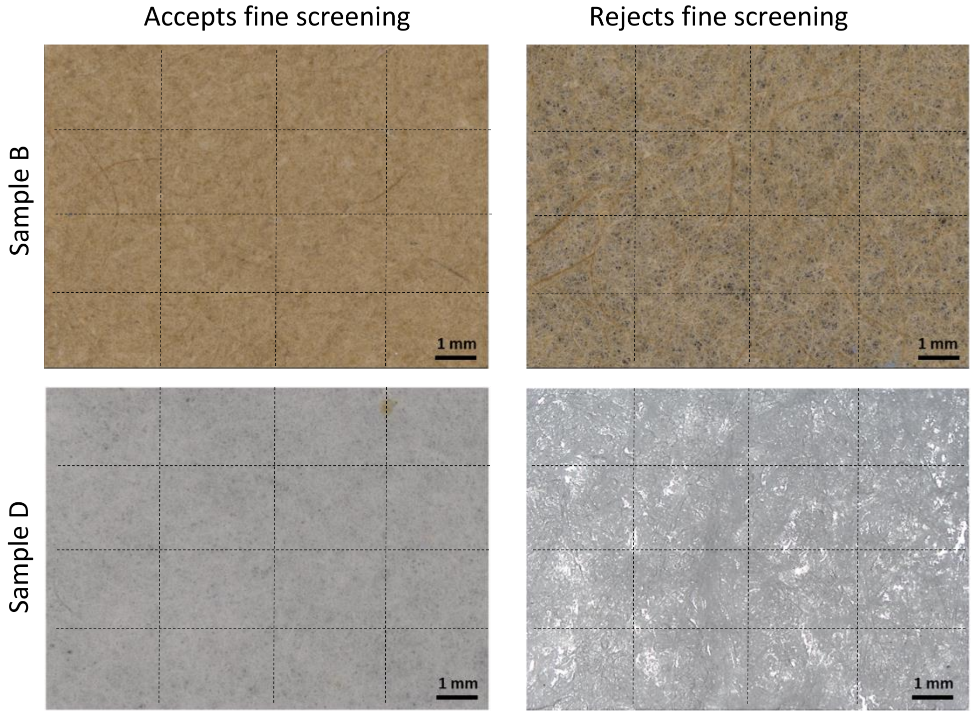

Figure 6. Large-area optical microscopy visualizing homogeneity of handsheets made from accept and reject fractions after fine screening for recyclable paper (sample B) and non-recyclable paper (sample D).

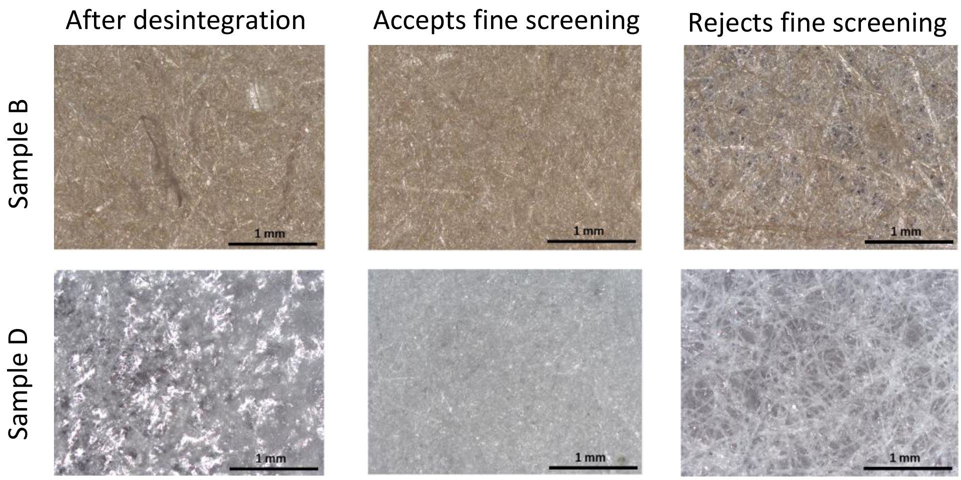

Figure 7. Detailed optical microscopy visualizing homogeneity of handsheets made from accept and reject fractions after fine screening for recyclable paper (sample B) and non-recyclable paper (sample D).

4 CONCLUSION

The laboratory-scale pilot line for recyclability testing of coated paper has successfully been validated for some industrial cases of coated packaging papers, resulting representative fiber yields recyclable versus non-recyclage papers. The repulping conditions are most sensitive to a threshold value for repulping time rather than repulping temperature. The coarse screening does not yield fiber rejects for recyclable coated papers, while it contains significant amount of polymer residues for non-recyclable papers. Alternatively, based on reference testing with soft tissue and newsprint paper, the coarse screening also separates a significant fraction of fiber agglomerates containing strong wet-end chemicals.

Acknowledgments: *This research was conducted as part of the CORNET project "REPAC²: Functional & recyclable coated paper packaging for food products". The funding for this research was provided by the Flemish Government through the grant number HBC.2021.0288, administered by the Agency for Innovation and Entrepreneurship (VLAIO-TETRA). We thank Patrick Doumen and Arne Derluyn for technical assistance.*

5 REFERENCES

Al-Gharrawi M.Z., Wang J., Bousfield D.W. (2021) Improving recycling of polyethylene-coated paperboard with a nanofibrillated cellulose layer. BioRes., 16, p. 3285.

Bilek M.A., Salem H., Korehei R., Olson J.A. (2021) Recycling Paper-Plastic laminate coffee cups using a Single-Disk Refiner: Energy requirements and recovered fiber quality. Waste Management, 136, p. 104.

CEPI (2022) Harmonised European laboratory test method to produce parameters enabling the assessment of the recyclability of paper and board products in standard paper and board recycling mills. Available through https://www.cepi.org/harmonised-european-laboratory-test-method-to-produce-parameters-enabling-the-assessment-of-the-recyclability-of-paper-and-board-products-in-standard-paper-and-board-recycling-mills (Accessed July 2023).

European Commission (2019) The green deal: sustainable industry. (2019) https://ec.europa.eu/commission/presscorner/api/files/attachment/859389/Sustainable_industry_nl.pdf.pdf

Fevia (2021) Verpakkingen 2025: onze engagementen. Accessed March 1, 2021. https://www.fevia.be/nl/verpakkingen2025-onze-engagementen

Kathuria A., Zhang S. (2022) Sustainable and repulpable barrier coatings for fiber-based materials for food packaging: a review. Front. Mater. 9, 929501.

Koppulu R., Lahti J., Abitbol T., Swerin A., Kuusipalo J., Toivakka M. (2019) Continuous processing of nanocellulose and polylactic acid into multilayer barrier coatings. ACS Appl. Mater. Interfac. 11, p. 11920.

Kunam P.K., Ramakanth D., Akhila K., Gaikwad K.K. (2022) Bio-based materials for barrier coatings on paper packaging. Biomass Conv. Bioref., 2022.

Lee K.S., Lee C.W., Han M.H., Ryu J.Y. (2020) Effects of recycling paper container for beverages on process water quality. J. Korea TAPPI, 52, p. 103.

Lee T.J., Yoon C., Ryu J.Y. (2017) A new potential paper resource: recyclability of paper cups coated with water-soluble polyacrylate-based polymer. Nord Pulp Paper Res J., 32, p. 155.

Ovaska S.S., Mielonen K., Lyytikainen J., Hirn U., Backfolk K. (2017) Determination of repulpability of talc-filled biopolymer dispersion coatings and optimization of repulped reject for improved material efficiency by tailoring coatings. PaperCon 2017 Paper conference.

Sridach W., Hodgson K.T., Nazhad M.M. (2006) Biodegradation and recycling of barrier coated paper. BioRes., 2, p. 179.

Tiagi P., Salem K.S., Hubbe M.A., Pal L. (2021) Advances in barrier coatings and film technologies for achieving sustainable packaging of food products – a review. Trends Food Sci. Technol., 115, p. 461.