Biodegradability and Recyclability of All-Cellulose Composite

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The development of sustainable packaging materials is becoming increasingly important to address environmental issues. In this context, all-cellulose composites (ACCs) represent a potential alternative to traditional composite materials. ACC consists entirely of cellulose and has significantly improved mechanical and barrier properties compared to paper. For packaging applications, the potential for material recycling and biodegradability are of great interest. However, these topics have so far been insufficiently addressed. In this study, the biodegradability of paper and different ACCs is compared. In addition, studies on the recyclability of the material were carried out. It was demonstrated that ACC is biodegradable. The rate of biodegradation depended on the parameters during the production process. Self-produced ACC was degraded at a rate almost identical to that of paper. The process takes longer for commercially available ACC. Furthermore, it was demonstrated that material recycling of ACC in the recycling paper loop was only feasible to a limited extent. The cellulose matrix between the fibres leads to a strong binding of the composite. This makes disintegration and fibre recovery difficult. Nevertheless, ACC represents a promising approach for packaging, particularly in regard to the global issue of littering.

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INTRODUCTION

Over the past decade, there has been a growing demand for more sustainable materials to replace plastic-based packaging within the industry. In this context, all-cellulose composites (ACCs) are a potential solution. ACCs were developed as early as the mid-19th century and have since been commercially distributed, for example, under the term *vulcanized fibre*. Numerous more recent studies have investigated novel starting materials and alternative production processes (Huber *et al.* 2012; Nishino and Peijs 2014; Fujisawa *et al.* 2018; Baghaei and Skrifvars 2020). In addition to the mechanical and barrier properties, the sustainability of ACC is frequently highlighted as a key advantage over conventional plastics and composites. This argument is based on two main factors, the use of renewable raw materials and the potential for biodegradability as an end-of-life strategy.

Biodegradability is particularly relevant for products that are at an increased risk of entering the environment after use. Naturally occurring cellulose is considered biodegradable and can be metabolized by various organisms, making it an attractive raw material for packaging applications from an environmental perspective (Eriksson *et al.* 1990). According to EN 13432 (2000), chemically unmodified materials of natural origin are considered biodegradable without testing. In comparison to naturally occurring cellulose-based materials, the crystal structure of ACC is modified. In addition to the naturally occurring cellulose I, cellulose II is present in ACC. Furthermore, the resulting materials often have a higher density than paper. The extent to which the formation of cellulose II affects biodegradability has only been discussed in a few studies.

An evaluation of the biodegradability of ACC was carried out by Kalka *et al.* (2014). For this purpose, ACC was produced from Rayon fibres using the solvent 1-butyl-3-methylimidazolium acetate. The biodegradability of the ACC samples produced was examined and compared with rayon fibre reinforced polylactic acid (PLA). For this purpose, samples of both materials were stored in a mixture of soil and compost for up to 70 days. Subsequently, a gravimetric determination of the remaining residues was conducted. For ACC, a mass loss of up to 73% was determined. Yang *et al.* (2010) and Zailuddin *et al.* (2020) used a comparable methodology and demonstrated that all-cellulose nanocomposites underwent substantial fragmentation within a few weeks, accompanied by a notable loss of mass. Another study evaluating the behavior of ACC in compost was conducted by Jahn *et al.* (2022). In this study, ACC was placed in a glass container and covered with compost. The fragmentation was visually assessed, revealing only slight differences between paper and ACC. However, the publications mentioned for the investigation of the biodegradation of ACC have a limitation, since only gravimetric or optical evaluation was examined; it is impossible, from such results, to distinguish whether the material has been metabolized and thus biodegraded or only fragmented into smaller components.

An accurate evaluation of biodegradation can be conducted by examining the released carbon or consumed oxygen associated with the metabolisation of carbon complexes. Wada *et al.* (2010) investigated the extent to which cellulose I and cellulose II can be converted into sugars by adding cellulase from *Trichoderma reesei* and cellulase from *Aspergillus niger*. The sample materials, microcrystalline cellulose (MCC) and bagasse, were analyzed in both their original state and after mercerization in an aqueous NaOH solution. The results demonstrated that the saccharification process was more rapid for the mercerized samples of MCC and bagasse in comparison to the untreated samples. Kobayashi *et al.* (2012) investigated the enzymatic degradation of diverse crystalline forms of cellulose. The saccharification of cellulose II was approximately 50% after 24 h when cellobiohydrolase from *Trichoderma longibrachiatum* was added. Miyaji *et al.* (2023) investigated the degradation of various forms of cellulose in natural waters. As part of the study, samples of cellulose powder, MCC, and a cellulose gel were analyzed. The oxygen demand during biodegradation was evaluated over a period of 30 days. The results showed that degradation occurred most rapidly in fresh water, followed by brackish water and seawater. After a period of 30 days, approximately 35 to 40% of the cellulose gel in fresh water had undergone degradation. Zhang *et al.* (1996) investigated the biodegradability of regenerated cellulose film; complete conversion of the cellulose film into water and carbon dioxide can occur within a period of two months. Park *et al.* (2004) examined the biodegradability of various textile semi-finished products based on cellulose and cellulose derivatives. After 24 days, 25 to 30% of the theoretically calculated total carbon of rayon

fibres was biodegraded, which is above the values observed for flax and cotton fibres. However, the study did not reveal the extent to which complete biodegradability can be achieved. The studies referenced demonstrate that cellulose II can be biodegraded with the addition of enzymes, as well as in natural waters. It can therefore be assumed that biodegradation in soil or compost is also possible. Whether this is the case, and if the degradation rates of paper and ACC are comparable, has not yet been investigated.

A preferred alternative to biodegradation is material recycling. It involves the utilization of products after their usage as raw material to produce new materials. For ACC only a few studies have been carried out investigating its recyclability. In a study by Spörl *et al.* (2018), ACC was produced *via* a two-step process using the ionic liquid 1-ethyl-3-methyl imidazolium acetate as solvent system. After production and material testing, the ACC was completely dissolved using the same solvent system. The resulting cellulose solution could then be utilized to produce new ACC. After three recycling cycles, only a slight decrease in the degree of polymerization was observed. No changes in the mechanical properties of the composite were detected. Accordingly, under ideal conditions, material recycling of ACC is possible. However, for an industrial realization, separate sorting and processing would have to be carried out. To what extent ACC can be used in already established recycling loops, for example for paper, is not known.

EXPERIMENTAL

Starting Materials

For papermaking, Northern Bleached Softwood Kraft (NBSK, Mercer International Inc., Berlin, Germany) composed of pine (40 to 70%) and spruce (30 to 60%) was used. The indicated percentages reflect typical variations in the supplier's production. All experiments were conducted using the same supplied pulp material, ensuring consistent fibre composition throughout the study. For the solvent system NaOH (99.6% purity) and urea (99.6% purity) were used and mixed with deionized water in the weight ratio 7:12:81 (NaOH:urea:H_2O) until a clear solution was obtained. Citric acid (food grade E330) was used to produce a 20% acid solution with deionized water, which is used for neutralization. Commercial ACC was provided by DYNOS GmbH.

Pulp Treatment and Paper Production

Pulp was treated in a Voith Lab Refiner LR40, using a universal beating disc with the parameters 3-1.6-60 and a specific edge load of 1,5 J/m. A 10-min machine warm-up period was implemented to prevent the formation of fibre lumps. Approximately 1500 g of pulp was used, with a moisture content of 7%. A beating energy of 100 kWh/t was applied. Dewaterability after refining is 27.0 SR (± 0.5 SR) (Schopper Riegler) according to ISO 5267-19 (1999).

Orthotropic sheets in accordance with ISO 5269-2 (2004) were produced from the refined pulp using a Rapid-Köthen Sheet Former. The laboratory sheets had a basis weight of 100 g/m² (± 2 g/m²).

All-Cellulose Composite Production

The conversion of paper to ACC was achieved through a 1-step process, using a custom-built laboratory unit. The system comprises an insulated and cooled area in which an immersion tank containing 20 L of solvent is situated. The solvent system and the

ambient air in this chamber were constantly kept at a temperature of -12.5 °C using a cooling unit and heat exchanger. The solvent was continuously stirred to prevent ice formation at the walls of the immersion tank and to maintain a homogeneous temperature distribution. Two additional dip tanks, situated outside the cooled area, were utilized for neutralization and therefore contained 20 L of 20% citric acid. The transfer of the sheets between the various tanks was carried out automatically.

During the conversion process, the paper sheets were initially immersed in the solvent system for a duration of either 1 s or 24 s. The sheets were transferred out of the solvent tank, where they remained within the cooled area for 1 s, allowing any residual solvent to drain off. The samples were transferred to the neutralization tank. Upon exiting the cooled area, a temperature increase from -12.5 °C to room temperature occurred. The transport times of 4 s were added to the immersion time, resulting in dissolution times of 6 s and 30 s. For neutralization, the solvent-soaked sheets were initially immersed in 20% citric acid for 1 min. Then they were stored in deionized water for 48 h. This was to prevent residues of the solvent or neutralizing agent from influencing the biodegradation. Finally, the ACCs were dried in a Rapid Köthen sheet former, according to ISO 5269-2 (2004).

Material Characterization

X-ray diffraction

X-ray diffraction (XRD) patterns were recorded on a Bruker D8 Advance Diffractometer operating at 40 kV and 40 mA with CuKα radiation and Bragg-Brentano geometry. The relative contributions of amorphous cellulose as well as cellulose I and cellulose II were estimated from the diffractograms. Due to the presence of broad and partially overlapping reflections, these values represent approximate estimations rather than precise quantitative phase fractions.

Aerobic biodegradation

The biodegradability test was conducted based on ISO 14851 (2019), an accepted method for assessing the aerobic biodegradation of natural and synthetic polymers. A two-phase closed bottle setup as described in Annex D of the standard was used. The incubation medium was standard nutrient solution and additional 5% (vol) of the supernatant of an activated sludge from a municipale sewage treating plant, without any preconditioning. Glass vessels (volume approx. 270 mL - the exact volume of each individual vessel V_v which is needed for calculation had priorly been assessed) were used as bioreactors. First, 200 mL of oxygen-saturated incubation medium (initial germ count: 8.0 x 10^4 cfu/mL) was filled into each vessel. The test materials were added to the vessels at a target concentration of 110 mg organic carbon/L. The test materials had been pulverized before using a standard kitchen appliance to minimize eventual impact of physical material characteristics such as thickness and density. The test was conducted in five replications per material, five blank vessels containing only the incubation medium were employed to determine background respiration. After measurement of initial dissolved oxygen concentration (WTW inoLab Oxi Level 2, equipped with a WTW CellOx® 325 probe), the vessels were sealed gas-tight with glass stoppers. This closed respirometric system was placed on a horizontal shaker for continuous homogenization. The incubation took place in the dark at 20 ± 1 °C; a pH between 6.5 and 7.5 was maintained throughout the whole period.

The concentration of dissolved oxygen in the incubation medium was measured at 31 dates during a total incubation time of 28 weeks, each first directly after opening the vessel (non-saturated state, c_{ns}) and a second time after re-saturating the incubation medium

by throughflow of ambient air (c_s). The vessels were sealed again directly after the second measurement.

Total oxygen capacity of the vessel (*i.e.*, oxygen in liquid phase plus oxygen in gas phase) in the saturated state at date *i* ($O_{s,i}$ (mg/vessel)) was calculated with Eq. 1,

$$O_{s,i} = c_{s,i}\, V_l + 0.28\, V_g \quad (1)$$

where 0.28 is the approximate oxygen content in ambient air (mg/mL), with barometric effects neglected, V_l is the volume of the liquid phase (200 mL), and V_g is the volume of the gas phase, calculated as individual vessel volume (mL) - V_l.

Total oxygen capacity in non-saturated state at date *i* was calculated based on Henry's law with Eq. 2.

$$O_{ns,i} = c_{ns,i}\, Vl + 0.28\, Vg\, c_{n,i} / c_{s,i} \quad (2)$$

Gross biological oxygen demand (*BOD*) was calculated for each individual vessel with Eq. 3.

$$BOD = \sum_{i=1}^{31} (O_{s,i} - O_{ns,i}) \quad (3)$$

To obtain net BOD for each material, mean BOD from the five blank vessels (5.3 ±1.6 mg) was subtracted from gross BOD values.

Conversion of BOD values into biodegradation was carried out by comparing measured net BOD with theoretical maximum oxygen demand (thOD), a value derived from elemental analysis of the tested materials (analyses for C, H, Cl, N, S, P, Na and O) and equation A.1 of Annex A of ISO 14851. ThOD of the four materials (paper, ACC 6 s, ACC 30 s, commercial ACC) accounted for 1.22, 1.20, 1.25 and 1.22 mg/mg, respectively. The incubation was stopped after 28 weeks as no significant difference between O_{ns} and O_s was measured anymore for any of the vessels (plateau phase).

Recyclability in paper loop

Recyclability *assessment* was based on PTS-RH 021/97 (PTS-RH 021/2012). This test specification is suitable for estimating whether paper and board products can be processed in paper recycling plants in such a way that they are suitable to produce new paper. The disintegrability of the paper or board is essential to recover the fibres. Additionally, the standard requires an investigation into whether additives included can cause the sheets produced from the recyclate to be sticky or to exhibit optical impurities. In the present case, however, it was known that the materials under investigation were free of additives, and thus only the disintegrability was analyzed.

To evaluate the disintegrability of the material, a representative quantity of the sample was cut into defined pieces and disintegrated under controlled conditions with the addition of water in a standard disintegrator, as described in ISO 5263-1 (2004). The standard disintegrating time was 20 min, which corresponds to 60,000 revolutions. Additional trials were conducted with a disintegrating time three times as long. The suspensions produced in this way were fractionated at a Brecht-Holl fractionator (ZELLCHEMING Merkblatt V/18/62) using a perforated screen with a hole diameter of 0.7 mm. Any residue remaining on the perforated screen was considered as reject, which was dried and weighed.

RESULTS AND DISCUSSION

X-Ray Diffraction

The X-ray diffractograms of the samples are presented in Fig. 1. For paper, typical peaks for amorphous cellulose and cellulose I dominated. Surprisingly, the results indicate that there was also a small amount of cellulose II. Similar values for paper were determined by Hildebrandt *et al.* (2017). For self produced ACC a slight change of the pattern between 15° and 17° indicates a reduction in cellulose I. However, there were no data showing the formation of cellulose II. In the *vulcanized fibre*, distinct changes were observed in the range of 22° and 34°, which indicated the formation of cellulose II.

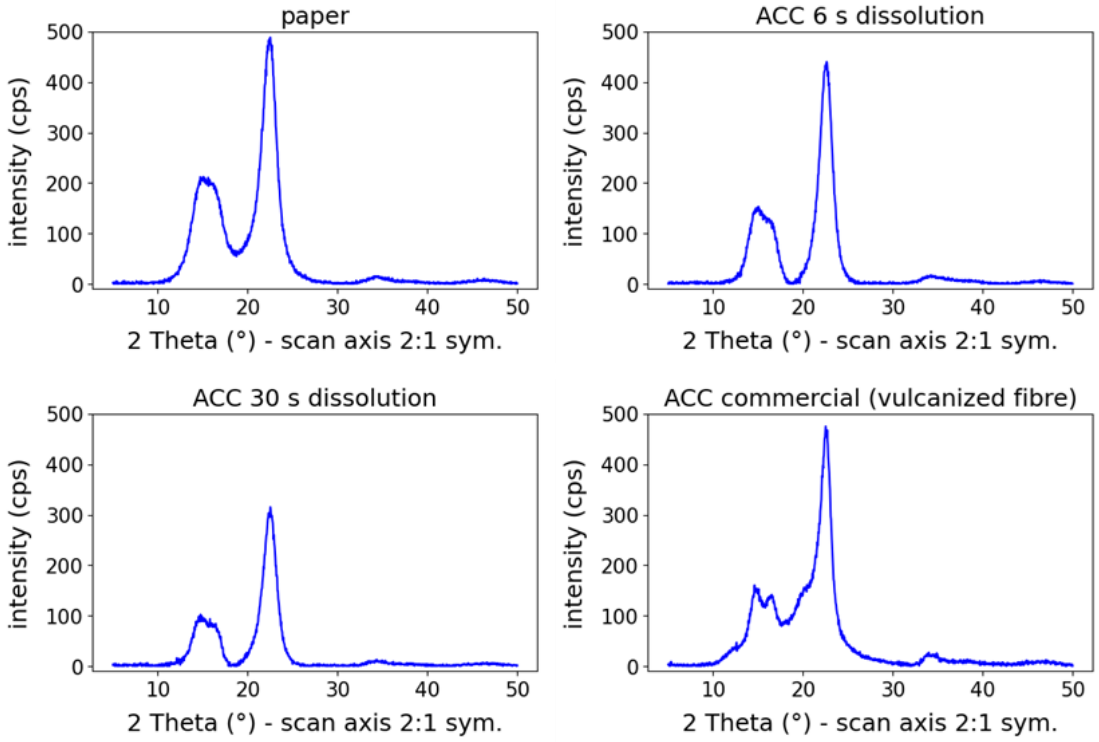

Fig. 1. X-ray diffractograms of paper (top left), self-produced ACC at a dissolution time of 6 s (top right) and 30 s (bottom left) and industrially produced vulcanized fibre (bottom right). Intensity is given in counts per second (cps).

The determined proportions of amorphous cellulose and cellulose I and II for the paper and vulcanized fibre are summarized in Table 1. It should be noted that the diffraction patterns exhibit broad reflections and partial peak overlap. Therefore, the reported phase contributions represent rough estimations based on relative peak intensities and do not constitute a full quantitative phase analysis. Interestingly, paper contained a small proportion of cellulose II.

Assuming that there is no cellulose II in natural occurring cellulose, this could be attributed either to an influence during pulp production or to measurement related deviations.

Table 1. Estimated Relative Contributions of Amorphous Cellulose, Cellulose I and Cellulose II

Sample	Amorphous Cellulose (%)	Cellulose I (%)	Cellulose II (%)
paper	6	89	4
self produced ACC 6 s	not quantitatively evaluated		
self produced ACC 30 s			
commercial ACC	6	69	25

Aerobic Biodegradation

Biodegradation reached mean values of > 92% for all materials, as summarized in Table 2. Single values of > 100% can occur due to measurement inaccuracies but also due to priming effects (Kuzyakov *et al.* 2000) and therefore are not unusual. Paper reached a mean of nearly 99%, suggesting a categoric difference between paper and all other materials rather than a correlation between ultimate biodegradation rate and degree of cellulose modification, but the difference between the rates observed for paper to the rates as calculated for the other materials is only partly close to significant, rendering this conclusion vague. The biodegradation rates reached at the plateau stage were within the specifications as defined by all relevant compostability standards concerning biodegradability.

Table 2. Gross BOD and Deduced Biodegradation Rates after 28 Weeks of Incubation (Means ± Standard Deviation)

	Gross BOD (mg)	Biodegradation (%)	Biodegradation relative to paper (%)
Paper	70.2 ± 3.8	98.9 ± 5.9	-
ACC 6s dissolution time	68.3 ± 1.5	94.7 ± 2.4	95.8
ACC 30s dissolution time	66.3 ± 1.8	92.3 ± 3.2	93.3
ACC commercial (*vulcanized fibre*)	64.2 ± 1.1	93.2 ± 2.7	94.2

More substantial statements can be deduced from the measurements if also the *dynamics* of biodegradation are considered. Figure 2 shows the biodegradation for all samples over a period of 28 weeks, as well as a fitted function from week 5 onwards. The fitted function was modelled using Eq. 4.

The biodegradation of both paper and the self-produced ACC show similar *dynamics*. In the first 2 to 3 weeks, intense biodegradation can be observed. Subsequently, the biodegradation rate decreases considerably until the fifth week, after which it follows a natural exponential function.

The observed *dynamics* in weeks 1 to 5 could be attributed to the initial degradation of more easily metabolizable carbon compounds, such as hemicellulose. After that, a decline in the rate of biological degradation is observed. From the fifth week onwards, the process aligns with the anticipated degradation *dynamics*. Initially, intense biological degradation can be observed. For longer incubation times, biological activity decreases again, which is due to the reduction in available carbon complexes.

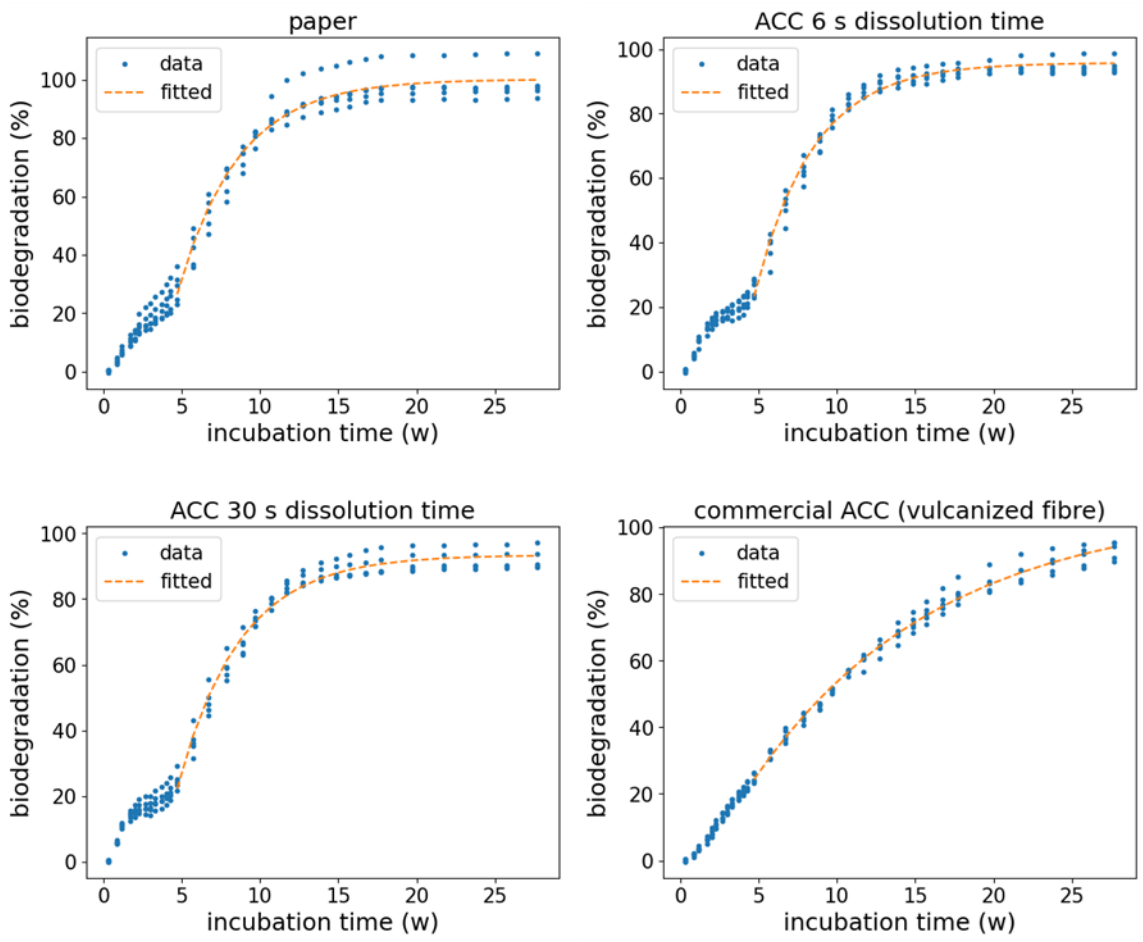

Fig. 2. Adapted aerobic biodegradation for a period of 28 weeks (w) and a fitted model (Eq. 4) for Paper (top left), ACC with a dissolution time of 6 s (top right), ACC with a dissolution time of 30 s (bottom left) and commercial available ACC (bottom right)

In the case of the commercially available ACC, a biodegradation that corresponds to an exponential function can be observed from the second week onwards. The starting product for commercial ACC is cotton linters, which has a very high content of alpha cellulose compared to NBSK (Sczostak 2010). This could be one reason for the lower biodegradation compared to the other samples in the first few weeks.

Based on the data shown in Fig. 2, models were created to describe biodegradation using an exponential function of the form, as follows,

$$y = A * exp^{(-B*t)} + C \quad (4)$$

where y represents the biodegradation, B the biodegradation rate and t the incubation time. A and B were included to compensate for an offset caused by deviations in the first 5 weeks of the experiment. For the modelling using a custom Python application, only the data from week 5 onwards were considered. The application uses the 'scipy' module and was run using Anaconda (Anaconda Software Distribution. 2020). The 'curve_fit' function, an integral component of the module, was utilized to calculate the parameters for Eq. 4, based on the experimental data. Table 2 summarizes the biodegradation rate (B) and the coefficient of determination (R^2) of the various materials.

Table 3. Model Parameters for-Biodegradation of Paper and ACC

	B	R^2
Paper	0.26	0.94
ACC 6s dissolution time	0.27	0.98
ACC 30 dissolution time	0.25	0.98
ACC commercial	0.08	0.99

Biodegradation rates were almost identical for paper and self-produced ACC. In contrast, the biodegradation rate of commercial ACC was significantly lower. XRD data showed that crystallinity and in particular the proportion of cellulose II in commercial ACC was significantly higher than in the other samples. Accordingly, crystalline cellulose I and cellulose II appeared to delay biodegradation. The determined biodegradation rate of over 90% for commercial ACC can be seen as proof that cellulose II is biodegraded under aerobic conditions. This interpretation is supported by XRD data indicating a clearly increased contribution of cellulose II-related reflections in this sample compared to the other materials.

Recyclability in Paper Cycle

Rejection rates determined during the sorting process for paper, self-produced ACC and commercially available ACC are summarized in Table 4. As expected, paper made merely from NBSK can be completely disintegrated, leaving no residual material during sorting process. For all ACC examined, the fibres could not be completely separated, leaving residues on the perforated plate, as shown in Fig. 3.

Table 4. Determined Reject Amount According to Paper and a Variation of ACC

Desintegration time (responding revolution)	20 min (60,000)	60 min (180,000)
Reject paper	0	not investigated
Reject ACC (6s Dissolution time)	50	25
Reject ACC (30s Dissolution time)	54	26
Reject commercial ACC (*vulcanized fibre*)	99	98

Fig. 3. Residues after desintegration (20 min) of paper (left) and ACC with a dissolution time of 30 s (right) on a Brecht-Holl-fractionator

Almost no disintegration occurred for commercial ACC. A tripling of disintegration time had no significant influence for commercial ACC. The determined amount of reject of ACC produced in this study corresponded to about 50% after a standardized disintegration time. A longer dissolution time during ACC production tended to have a negative influence on disintegration. However, further tests would be necessary for a statistical evaluation. A tripling of the disintegration time resulted in a reduction in the amount of reject.

According to the PTS method RH 021/12, samples with residues between 20% and 50% are classified as recyclable, but in need of improvement in terms of product design. Accordingly, the ACC produced during this study was on the very edge of limited recyclability. The tested commercially produced ACC samples were classified as non-recyclable. In a modern paper mill, screening equipment routinely removes flakes of recovered paper (as shown in Fig. 3) that otherwise would create blemishes in the recycled paper. However, the rejected material, if not recovered by specialized disintegration procedures, will decrease the yield of the process.

It was expected that ACC would be more difficult to disintegrate due to the compaction of the material and the high wet strength compared to the base paper. The energy applied during disintegration was therefore not sufficient to completely break the fibre bonds and the cellulose matrix between the fibres. Even when the disintegration time was tripled, the ACC could not be completely disintegrated. According to the current state of knowledge, material recycling of ACC in the established recycling paper loop is only possible to a limited extent. However, the results show that the manufacturing conditions could have a significant influence on recyclability. Laboratory sheets formed from the accept fraction showed no sticky contaminants or inhomogeneities, indicating that the recovered fibers themselves are processable once separated.

CONCLUSIONS

1. Under the condition that no cellulose derivatives were present and that all processing chemicals had been completely removed, the all-cellulose composite (ACC) was found to be fully biodegradable under aerobic conditions.
2. The biodegradation rates of ACC were lower than those of paper and could be influenced by the manufacturing process and the starting material.
3. The conversion of paper to ACC led to the formation of a cellulose matrix and increased structural integrity, which significantly hindered fibre separation during disintegration and reduces recyclability. Controlling the degree of partial dissolution was found to be a key strategy to balance mechanical performance and fibre recoverability.
4. Recyclability of ACC in the recycling paper loop was found to be only possible to a limited extent and must be evaluated for each individual product.
5. In the specific case, commercially available ACC could not be disintegrated. ACC produced in this study had a limited recyclability, depending on the manufacturing conditions. In a modern paper recycling facility this is expected to result in a lower yield.

6. The mechanical properties and reprocessing performance of fibers recovered from ACC require systematic evaluation, particularly with respect to strength development, bonding ability, and process stability in repeated recycling cycles.

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