

Biobased polymers for Meltblown Nonwovens

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The development of bio-based alternatives to conventional olefin-based plastics represents a major focus of current research. However, many bio-based polymers remain limited in terms of processability across different processing technologies and in their application performance. While Polylactic acid (PLA) has been successfully established on an industrial scale over the last 20 years, polyethylene furanoate (PEF) has emerged as a promising biobased alternative or complement to polyethylene terephthalate (PET) in various applications such as food packaging, bottles and technical as well as high-performance fibers. Further, polyhydroxyalkanoates (PHAs) have been focussed, although they are still limited to injection molded and/or electrospun applications.

In our studies we successfully demonstrated the processing of PLA, PEF and also PHA nonwovens via meltblow technology. PEF was shown to be equivalent or even superior compared to PET and Polybutylterephthalate (PBT) fabrics of similar grammage concerning achievable fiber diameters, mechanical strength and filtration performance. PEF as well as PLA can thus be considered as biobased alternatives, particularly for applications requiring thin layer geometries at elevated temperatures. In addition, the potential of PHAs, especially poly(3-hydroxybutyrate) (P(3)HB) has been demonstrated across a range of applications. Nonwoven structures with fiber diameter distributions ranging from 2.5 μm to 20 μm could be laid down, exceeding the shortcomings of high brittleness, low elongations at break (max. 3%) and a lack of flexibility using the copolymer poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBH). Furthermore, the high melt adhesion and the special crystallization kinetics of PHAs have commonly been regarded as constraints in filament and nonwoven processing so far. However, these two properties have now been used to develop a three-dimensional fiber arrangement, resulting in dimensionally and temperature-stable fabrics. These nonwovens offer an interesting array of properties by combining moderate gas permeability and hydrophobicity, leading to water (lq.) impermeability.

In summary, PLA and PEF exhibit high potential for applications in particle and dust separation. In contrast, PHA, particularly PHBH, have been identified as suitable materials for gas-water or liquid separation processes, as well as for use as bio-based and biodegradable carrier systems.

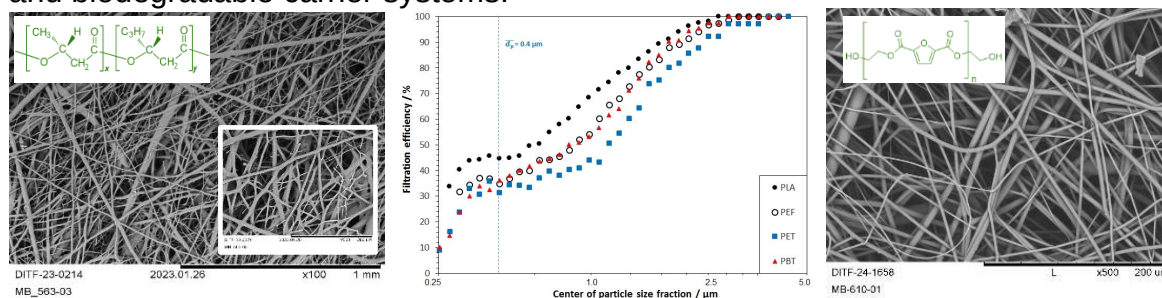


Figure: REM images of PHBH (left) and PEF nonwovens (right) and comparison of filtration performance compared to reference materials (center)

Keywords: Biopolymers, Biodegradables, Fibers, Nonwovens, Meltblown, Filtration

Introduction

The meltblow process represents an efficient technique for the direct formation of nonwoven webs composed of finest fibers from the polymer melt. Due to the resulting fiber fineness, high porosity and correspondingly small pore size distribution, meltblown nonwovens are particularly well suited for filtration applications. In the most widely applied and industrially established Exxon-type die configuration, the polymer melt stream exits a prismatic die assembly comprising several hundred linear capillaries and is immediately captured by two converging, high-velocity laminar streams of heated primary process air. However, this die design exhibits limited pressure resistance, thereby significantly reducing the range of processable polymers. According to IUPAC, the term “bioplastics” corresponds to “bio-based polymers” and refers to polymers on the basis of “biomass or monomers derived from biomass [...]” [1]. In line with the rising awareness for the future need for fossil-free plastics, the search for bio-based monomer sources or bio-based polymers with equal or at least close to equal properties is rapidly increasing to provide a feasible alternative for fossil-based polymers. Among a variety of different polymer types PLA is the most frequently reported and most industrialized member of the family of bio-based polymers. Despite a limited biodegradability even in industrial composting [2], PLA-based nonwovens may offer advantages over other biobased nonwovens in terms of production costs, commercial availability and application-specific properties such as flexibility and thermal resistance. Notably, desirable meltblown fiber characteristics and corresponding nonwoven performances have already been demonstrated [3,4]. Recently, PEF has been the focus of intensive research as a bio-based alternative to PET with the potential to partially or fully substitute PET in existing applications and products. The interest arises from the chemical similarity between the two polymers. Yet while PEF can potentially be produced entirely from renewable resources, PET can only achieve a maximum of 50% bio-based content. Furthermore, PHAs represent a promising class of polymers that combine bio-based origin with biodegradability under various environmental conditions. Although more than 100 different PHA variants have been identified to date, industrial relevance remains concentrated on poly(3-hydroxybutyrate) (P3HB) and some few select copolymers (mainly poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) and more recently poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBH)), due to the polymer price and the available scale. Notably, to date a wide range of biotechnological and synthetic approaches for the production of those bio-polyesters has been established.

PLA: Competitive against commodity polymers on industrial scale

Among other bio-based polymers, PLA is the most developed and the most industrially established polymer. This benefits PLA-based nonwovens compared to other bio-based or biodegradable polymers in terms of costs, market availability, but for the current state also in application-specific properties like the bending behaviour or thermal resistance, which can compete with established fossil-based polymers like polyethylene (PE), polypropylene (PP) or PET. One further advantage to polyolefins is that PLA is biodegradable under industrial conditions.

Previous research has focused on the production of fine-fiber PLA webs, achieving fiber diameters as small as 0.5 μm [3]. However, they were produced under specialized equipment and laboratory conditions. Other studies explored the influence of

processing parameters on fiber diameter distribution, pore structure, and filtration efficiency [5,6]. It was demonstrated that electret charged PLA fabrics can achieve filtration performance comparable to PP meltblown.

Based on the preliminary work, our results showed that PLA can be processed into high-performance meltblown filtration materials under industrially relevant Exxon-type conditions [7]. When processed under identical specifications and at the same production scale, PLA can achieve fiber diameters, nonwoven structures, and functional properties comparable to those of PP. Furthermore, PLA demonstrates a pronounced sensitivity to variations in processing parameters, enabling the controlled formation of ultrafine fibers typical for meltblown nonwovens and coarser, spunbond-like fibers, as exemplarily shown for two different process settings of the same PLA grade in **Figure 1**.

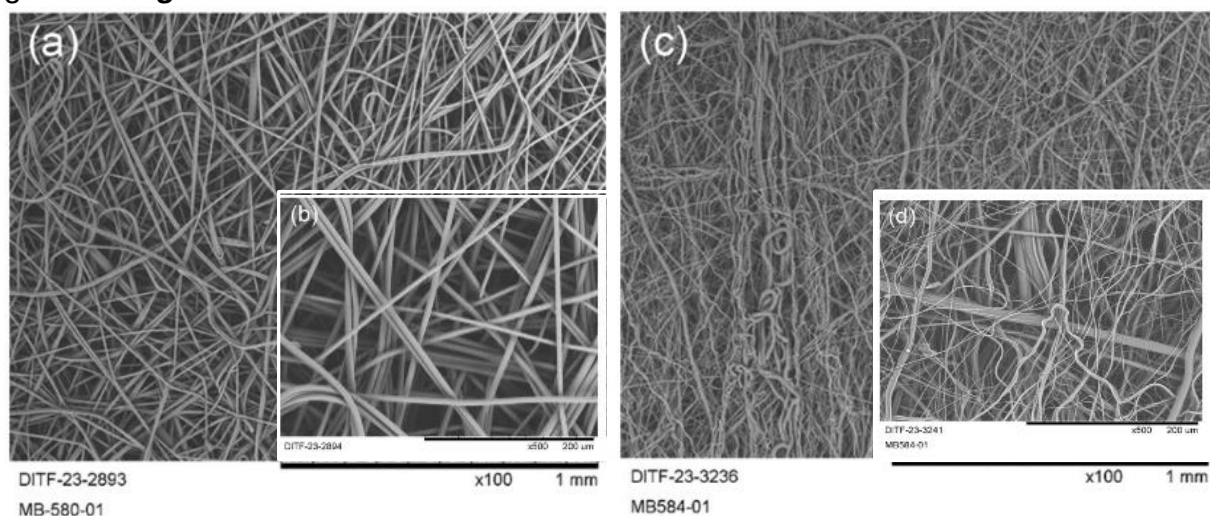


Figure 1: SEM images: (a) PLA-meltblown (coarse); $\times 100$, (b) PLA-meltblow (coarse), (c) PLA-meltblow (fine); $\times 100$, (d) PLA-meltblown (fine).

The fiber diameter distribution evaluated from the images shows **Figure 2**.

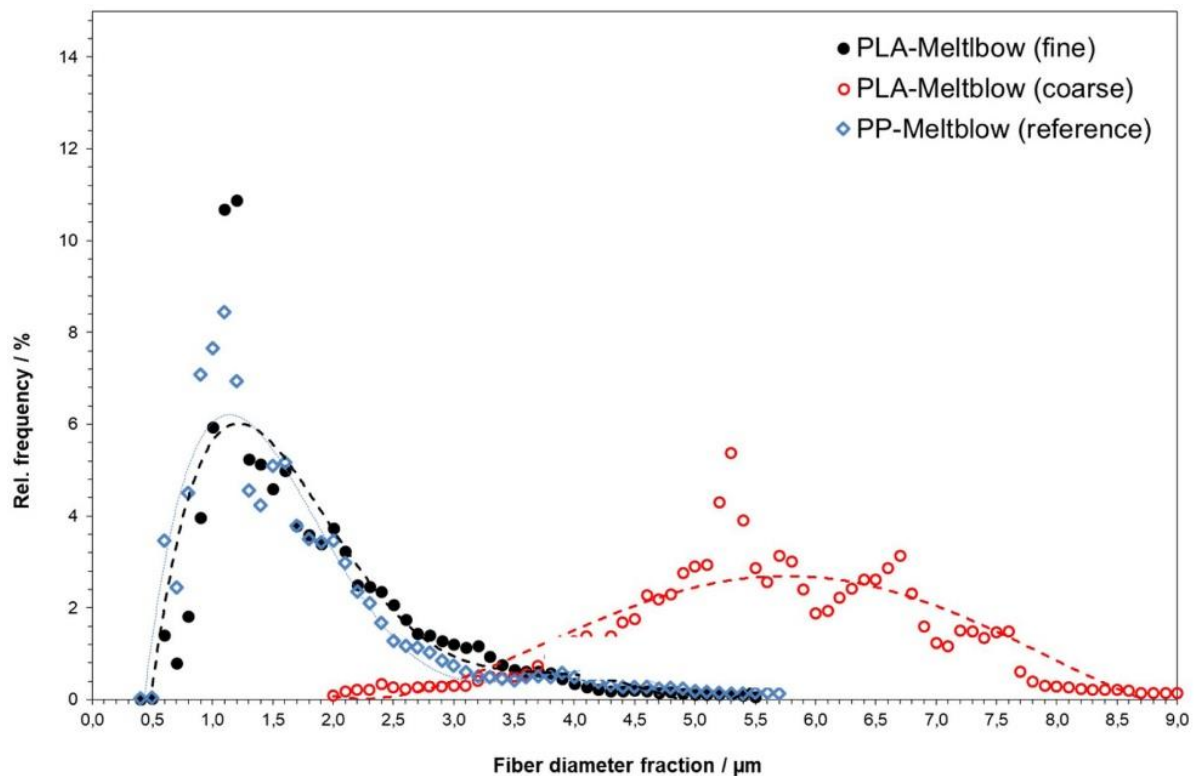


Figure 2: Fiber diameter distributions (from SEM, $\times 1000$) of PLA meltblown nonwoven samples and PP reference.

The results indicate that fine fiber fabrics produced using PLA, exhibited a comparable diameter to those made from PP (Median diameters: PLA: $1.37 \mu\text{m}$ vs. PP $1.41 \mu\text{m}$). Similarly, the resulting fabric thickness ($\sim 90 \mu\text{m}$ according to DIN EN ISO 9073-2) and air permeability values ($\sim 1000 \text{ l m}^{-2} \text{ s}^{-1}$ according to EN ISO 9237) lie within the same range. This indicates that the pore sizes and also the specific surfaces are directly comparable. However, the filtering effect¹ was more efficient with PP achieving 90% ($0.25 - 4.0 \mu\text{m}$ particle size range) and also lower pressure drop of 27 Pa compared to PLA nonwovens. Notably, the filtration efficiency of the PLA nonwoven in our studies, reaching 75% at 38 Pa, was higher as described for comparable PLA-nonwovens in former studies [5,6]. As the influence of specific surface area was previously excluded, one plausible explanation may be a tribological effect involving friction and charge carrier transport along the fiber surface. Addressing this aspect, a key finding of the present study was that electret charging² increased the aerosol/particle filtration efficiency of fine-fiber PLA meltblown nonwovens (grammage: 25 g m^{-2}) to approximately 99%, thereby matching or surpassing the performance of PP materials with comparable fiber diameter distributions. Furthermore, the electret-induced filtration performance remained above 90% over several weeks, demonstrating the temporal stability of charge retention.

¹ measured in reference to ISO 16890 following the standard for filtering half mask classification (FFP according to EN 149) with adapted test parameters to a test area of 100 cm^2 (flat, circular rondes), volume flow of 95 l min^{-1} , and a flow velocity of 9.3 cm s^{-1} using liquid paraffin oil (density $0.825\text{--}0.850 \text{ g cm}^{-3}$ at room temperature)

² The fibers were charged online in the meltblow process by passing two electrodes applying a voltage of + and - 30 kV DC.

Summarizing, the effect of process parameters on the meltblown, results are illustrated in **Figure 3**.

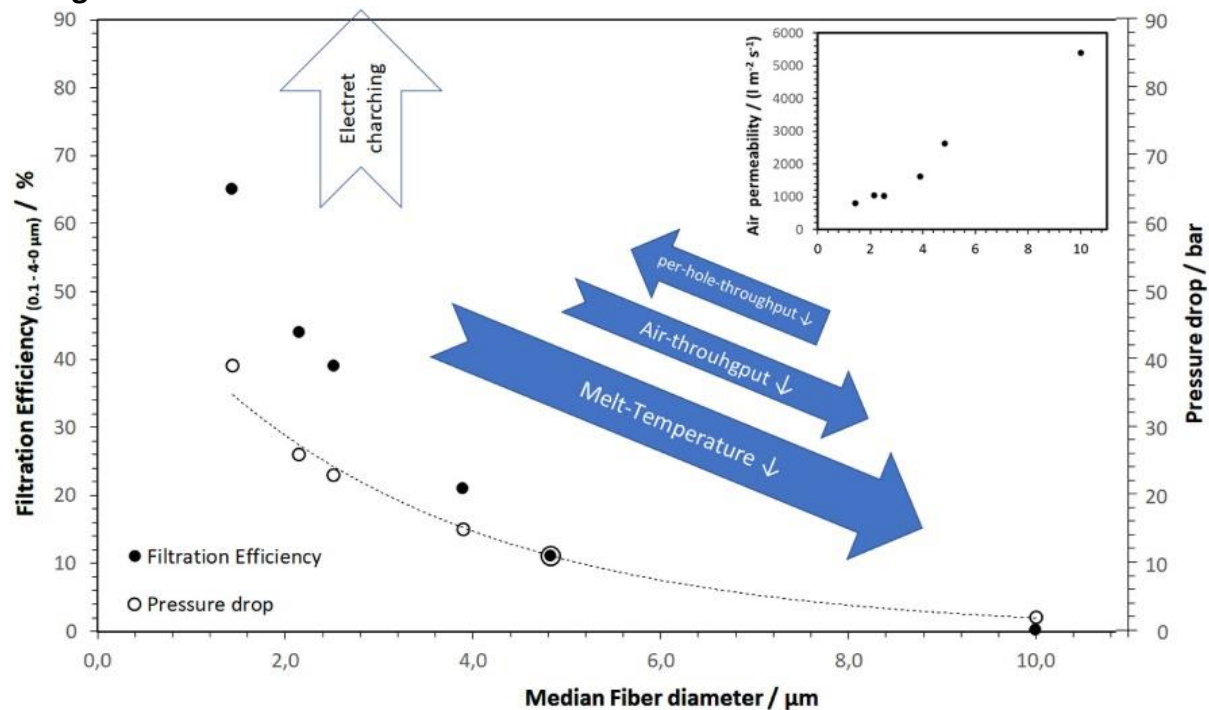


Figure 3: Effect of meltblow process parameters on filtration efficiency and pressure drop variation as a function of the median fiber diameter.

With increasing melt viscosity, resulting from lower temperatures, and a reduced amount of process air (lower acting forces) thicker fiber diameters and more open nonwoven structures with larger pore sizes are formed. This effect can be further enhanced by increasing the throughput per capillary, and vice versa. Although coarse nonwovens (median fiber diameter: $\sim 6 \mu\text{m}$) still exhibit inferior mechanical strength compared to consolidated PLA spunbond materials, these findings highlight the potential to produce multifunctional carrier – filtration layer – media only using the meltblow technology. A promising approach for further research is the post treatment of the deposited nonwovens, for example by systematic calendaring, in order to enhance mechanical strength and achieve strength levels comparable to spunbond media.

Overall, this research contributes to identifying the potentials of replacing fossil-based PP in filtration applications with bio-based PLA. Consequently, PLA emerges as a strong candidate for the future development of sustainable nonwoven technologies, particularly in air filtration, medical masks, and lightweight composite systems.

PEF: 100% bio-based polyester

In recent years, PEF has been identified as a bio-based alternative or supplement to PET for various applications such as food packaging, bottles, and technical- as well as high-performance fibers and yarns [8].

Both, PEF and PET, are synthesized through melt polycondensation of ethandiol (with a dicarboxylic acid). Meanwhile, ethandiol can be gained on biological basis, potentially leading to PET, which is 50% bio-based. However, 2,5-Furan dicarboxylic acid (FDCA), can be extracted on bio-chemical routes from agricultural waste streams like

chicorée routes. FDCA is chemically similar to terephthalic acid (PTA), the second building block of PET, which is shown in **Figure 4**, substituting the phenylene ring by a furan ring.

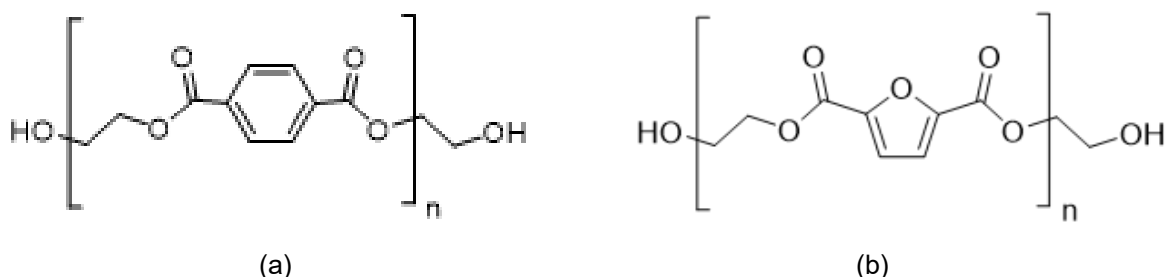


Figure 4: Chemical structures of (a) PET and (b) PEF.

Using PEF, two key goals of Germany’s “2030 National Research Strategy for BioEconomy” are fulfilled: “Industrial usage of renewable resources [...] neither additional agricultural land is required, nor crude oil is consumed”. [9] Comparing the chemical structures of PEF and PET, the substitution of the phenyl ring by a furan ring, leads to reduction of chain mobility due to the non-linear structure and a more rigid polymer backbone. Due to this PEF is superior to PET in some aspects, for example higher gas impermeability to carbon dioxide, oxygen and water vapor [10] and a higher glass transition temperature (T_g) (85 – 89 °C vs. 70 – 76 °C) combined with a lower melting range (~210 – 215 °C vs. 250 – 260 °C) [11]. Although PEF exhibits slower crystallization kinetics and a reduced tendency for the development of oriented amorphous phases compared to PET [12], this behaviour poses challenges for various processing technologies.

Besides these challenges, we could demonstrate, that PEF is compatible to PET in all aspects of the meltblown process. The results demonstrate the feasibility of achieving industrially relevant per-hole throughputs (> 0.1 g/hole/min) while maintaining fine (micro) fiber diameter distributions (2.0 µm in median). The resulting fibers exhibit mechanical properties (in both machine and cross direction) and air permeability values comparable to those of PET. Furthermore, a notable filtration performance was achieved, with efficiencies exceeding 50%³ (35% for the 0.4 µm fraction) at a pressure drop of 19.5 Pa for nonwovens with a grammage of 25 g m⁻² prior to electret treatment. Following electret charging the filtration efficiency increased to above 90 % after electret treatment (85% for 0.4 µm fraction) at a pressure drop of 18.3 Pa.

A comparison of the filtration curves of PEF with those of PET, poly(butylene terephthalate) (PBT),⁴ and PLA is presented in **Figure 5**.

³ Particle size range from 0.2 µm to 4.0 µm

⁴ Most common used polyester type in meltblown showing fast crystallization kinetics.

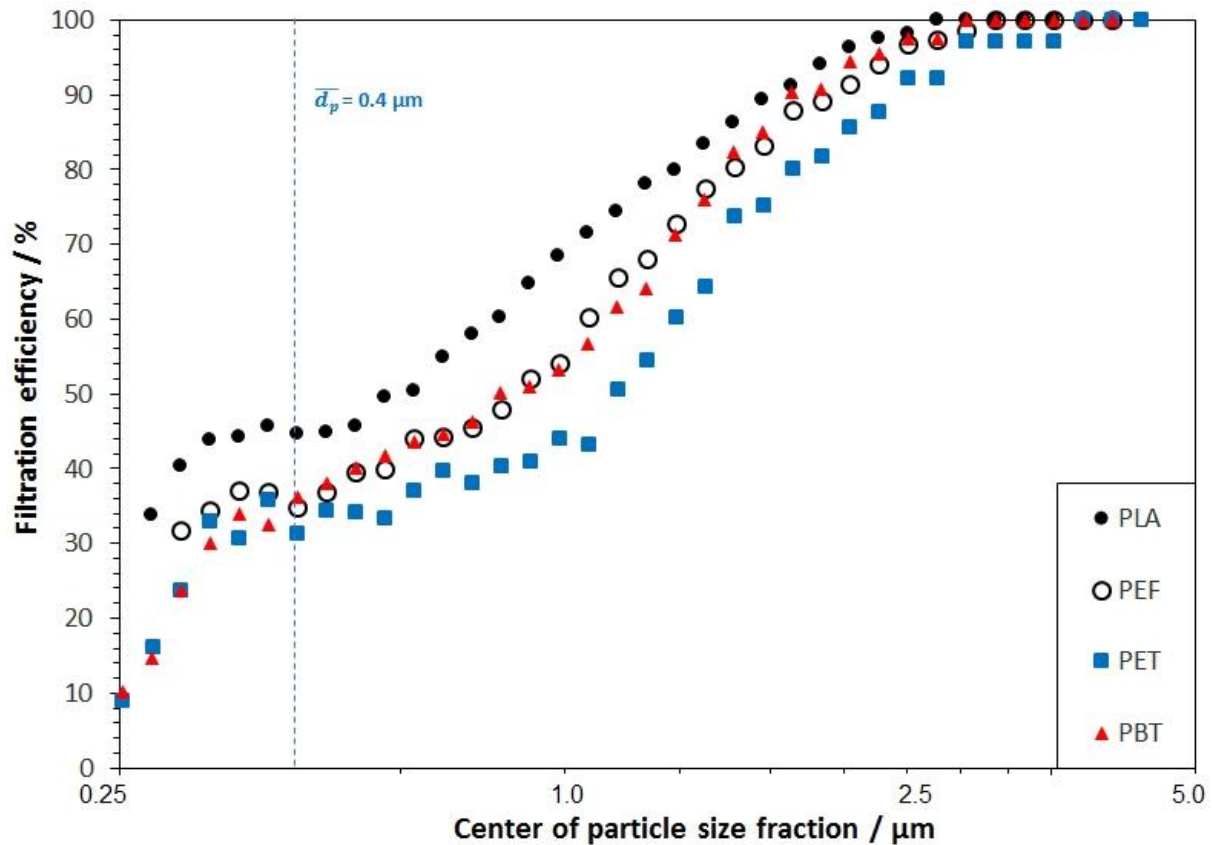


Figure 5: Fractional particle filtration efficiency against paraffine oil (9.3 m s^{-1}) for PLA, PEF, PET and PBT meltblown fabrics with a grammage of 25 g m^{-2} .

Indeed, benefits of PEF compared to PET could be revealed such as a lower processing temperature (energy consumption) towards PET (270°C vs. 280°C of melt and 285°C vs. 300°C of air). One aspect that still requires improvement for PEF compared to established polyesters is its thermal stability, as indicated by thermal shrinkage of 20 - 30% after exposure to 200°C for 15 min. However, this limitation may be addressed by implementing additional post-heating zones between fiber laydown and winding. Owing to the comparatively slow crystallization kinetics of PEF, such process adaptations can enhance crystallinity and thereby improve thermal stability. Wide-angle X-ray scattering (WAXS) and differential scanning Calorimetry (DSC) analyses revealed that the fiber crystallinity of PEF is around 7%, whereas PET and PBT reached 27% and 38%, respectively, under comparable process throughputs on the same experimental setup. This is below the minimum crystallinity of 25% required to prevent thermal shrinkage [13]

The current development status of PEF fibers and nonwovens indicates that their properties have high potential for diverse applications. The main hurdle to further optimize the thermal behaviour of PEF is the cost of raw materials as well as the availability in polymerizable quality, rather than technical feasibility. If the polymer price develops similar to what was seen with PLA, which is necessary to enable PEF entering the industrial sector, PEF could be established with a focus on bio-based sustainability and new accessible properties.

PHAs: 100% bio-based polyester

The term of bioplastics or biopolymers is widely used imprecisely. Biodegradable polymers (which decompose under certain conditions only into CO₂ and water as residues) are not necessarily also biobased (polymers on the basis of biomass or monomers derived from biomass) and vice versa. A promising material class, combining both properties, are PHAs. There is a variety of biotechnological and synthetic approaches to produce these bio-polyesters. Among others, they can be gained by biosynthesis in natural isolates, genetically engineered plants, different recombinant bacteria and various types of archaea [14,15]. These organisms produce linear polyesters to store energy and carbon by intracellular inclusions [15]. Although more than 100 different PHA variants are known to date, the industrial relevance still lies with P(3)HB and some selected copolymers mainly PHBV, more recently PHBH, due to the polymer price and the available quantity. For meltprocessing of fibers, one main challenge is, that their crystallite melting temperature is close to the degradation onset temperature. Further limitations are factors such as a high melt adhesion, a low melt strength and extensibility, as well as a low initial crystallization rate in conjunction with strong post (secondary) crystallization. For P(3)HB, the latter leads to high degrees of crystallinity up to 80 % with a large spherulite size and thus to a high brittleness, which is disadvantageous for industrial use.

Experimental meltblown trials using different industrial PHAs, combined with comprehensive material and nonwoven analyses, revealed the requirements for a successful and stable processing on both the material and process side beyond laboratory scale, and in the absence of additives. It was demonstrated that the basic processability in terms of extrusion and fiber formation at the spinneret can be estimated by the complex shear viscosity by measuring the static absolute value of complex viscosity over time (at constant shear and strain). It was found that polymer processability improves with a broad molar mass distribution (>8), with more and high molar mass fractions ($>10^6$ g·mol⁻¹) are employed, as this contributed to enhanced processing stability by balancing the thermal decomposition process. The shortcoming of PHAs such as post-crystallization, which was still present in low molar mass grades with narrow distribution, was additionally overcome by the higher molar mass, and the benefits of low molar mass fractions for melt flowability were maintained. In addition, degradation was found to be rather counterproductive at narrow molar mass distributions, resulting in an unstable process as well as reduced process variability concerning productivity and variability of fiber diameter formation.

By meltblowing, fiber diameters below 10 µm were achieved [16], while the melt adhesion could be suppressed by employing process air slightly cooler than the melt temperature. Furthermore, the embrittlement of the resulting P3HB nonwovens was mitigated through the use of selected PHBH grades. [17]

Median fiber diameters with a minimum of 2.4 µm were achieved, which is remarkable compared to values reported in literature (>14 µm) [18]. Moreover, the median

diameter could be adjusted up to 20 μm , allowing for tunable fiber diameters suitable for different applications.

One special aspect of PHA was found in the potential to form and obtain time and temperature stable three-dimensional fabrics based on the hypothesis of combining the high melt adhesion and the special crystallization kinetics - two properties of PHAs that have so far only been considered as limitations in filament and nonwoven processing. Using these two properties as benefits in the meltblown process, three-dimensional objects were produced directly on the conveyor belt (scheme shown in **Figure 6**).

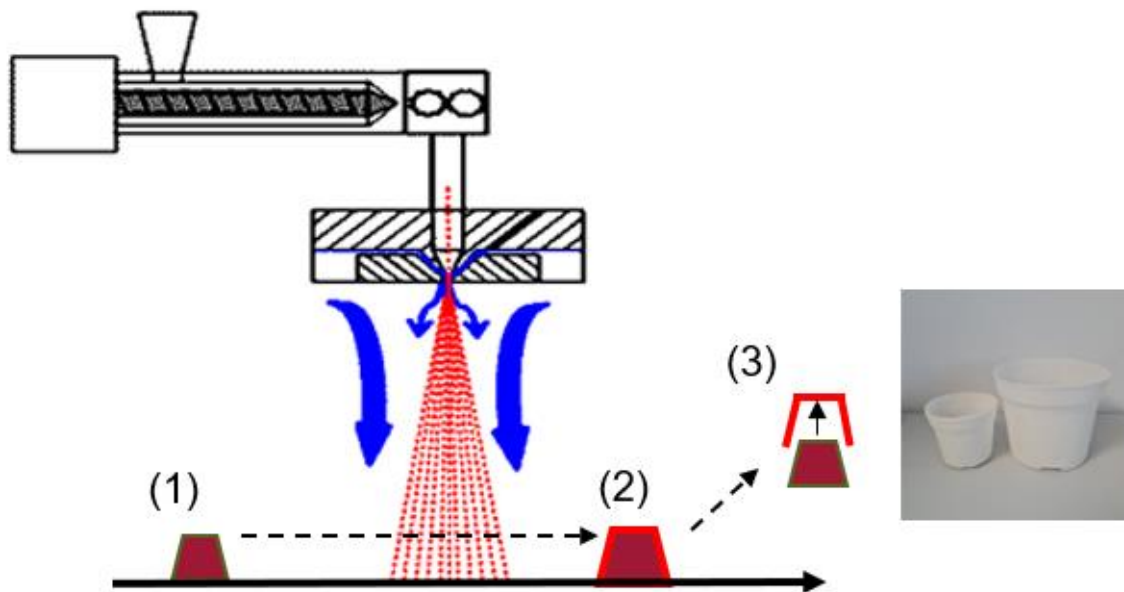


Figure 6: Concept for the production of three-dimensional meltblown PHA-"fabrics".

A planting pot was chosen as a demonstrator, as a simple but application-relevant object shape. The pots produced were characterized and compared with commercially available reference materials. It was shown that the shape of three-dimensional nonwovens made of PHBH was completely elastic in compression, in contrast to PHB and PHBV. A further interesting constellation of properties results in moderate air permeability and hydrophobicity, which leads to impermeability to water. Although the particle filtration efficiency remains still low, due to the comparable coarse fiber diameters, see **Figure 7**, the findings open up a wide range of applications for novel nonwovens and three-dimensional shapes made from this biobased and biodegradable polymer family of PHA, e.g. in medical applications.

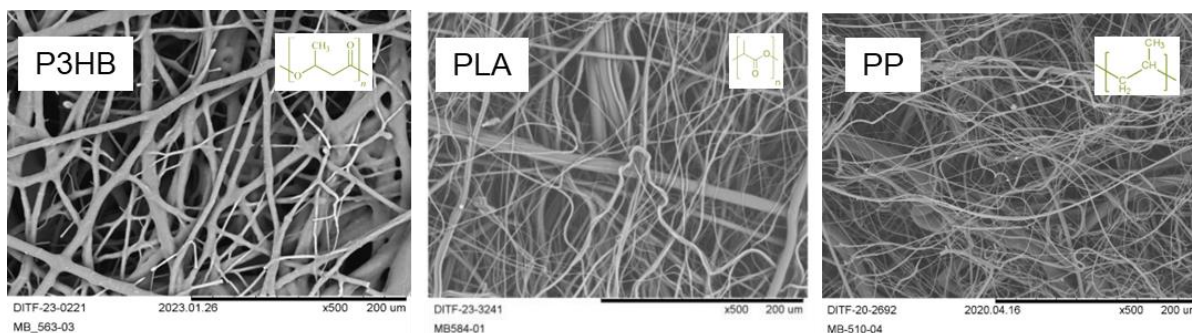


Figure 7: Comparison of the fiber deposition of P(3)HB, PLA and PP in Scanning Electron Microscope (SEM).

Conclusion

Meltblown nonwovens of bio-based polymers exhibit high potential to compete with conventional fossil-based materials. However, primarily due to current polymer costs, individual application-specific aspects have to be carried out in order to supplement or fully replace established polymers. Thus, the relevance of biodegradability must be evaluated in the context of the intended application and environmental conditions, as it may be advantageous or detrimental. Furthermore, the CO₂-footprint, accounting for the polymer's bio-based origin, should be considered, an aspect that has been largely neglected in the case of PLA for a long time [19]. However, PLA is the most established bio-based polymer and (already) market available in suitable quantity. This has to be the reference for other bio-polymers for transferring promising potentials into industrializable approaches.

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