



# Exploring polyethylene/polypropylene nonwoven fabrics derived from two-dimensionally co-extruded composites: Effects of delamination, consolidation, drawing and nanoparticle incorporation on mechanics, pore size and permeability

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## ARTICLE INFO

### Keywords:

Coextruded composite  
Drawing  
Consolidation  
Nonwoven fabric (mat)  
PMMA colloidal nanoparticle

## ABSTRACT

Co-extruded fiber-like composites after delamination facilitate the synchronous production of dual component fibers. In this study, high-density polyethylene (HDPE)/polypropylene (PP)/polystyrene (PS) and low-density polyethylene (LDPE)/PP/PS composites (fiber precursor) were melt-processed by using multilayer coextrusion. Each composite was uniaxially drawn to enhance mechanical robustness of HDPE, LDPE and PP fiber-like domains. Following the drawing process, the sacrificial PS layer was removed, and a combination of hydroentanglement, needle-punching, and compression molding was utilized to formulate dual-component HDPE/PP and LDPE/PP nonwoven mats. Drawing of the coextruded fiber precursor enhanced the tensile strength of the HDPE/PP nonwoven mat from 5.5 to 56 MPa up to a draw ratio of 11. To decrease the pore size, in the dual-component nonwoven mat, nanoscale poly (methyl methacrylate) (PMMA) colloids were infused into the large pores comprising the void spaces within the nonwoven fabric, thereby effectively reducing the pore size from 10.7 to 0.8  $\mu\text{m}$ . By using the dual-component fiber system, pore size can be reduced by post-processing at a temperature between the melting points of the two different fiber components. Due to that, the efficacy of these nonwoven mats as a physical separator in battery applications was assessed via thermal shutdown analysis.

## 1. Introduction

Polyolefins such as low-density polyethylene (LDPE), high-density polyethylene (HDPE), and polypropylene (PP) have been widely used in various applications due to their excellent chemical resistance, mechanical properties and low cost [1–4]. Electrospinning has recently garnered a great deal of attention with the advent of drawing aligned fibers, capability to easily achieve sub-micron diameter fibers, and creation of a nonwoven fiber mat without additional post-processing [5–7]. However, conventional electrospinning requires high temperature and harmful organic solvents in order to process polyolefin fibers [6,8]. As a result, melt-spinning and melt-blowing are the most common methods to process chemically resistant polyolefins into high value-added non-woven fabric mats [9–12]. When consolidated into a nonwoven fabric, polyolefins exhibit considerable potential in a wide range of fiber applications due to their porous structure, flexibility, and low cost in particular. However, these manufacturing methods allow for

a limited draw ratio of fibers when oriented [13–15].

Recent work utilizing multilayer coextrusion has demonstrated the production of rectangular fibers with lateral dimensions on the sub-micron scale for systems of LDPE, HDPE, PP, poly ( $\epsilon$ -caprolactone) (PCL), polyamide-6 (PA6) and poly (ethylene terephthalate) (PET) [16–18]. The delaminated co-extruded polyolefin fibers are particularly advantageous owing to the flexible nature of the coextrusion system and low material cost [16]. In spite of the potential uses, studies on consolidation of these novel co-extruded fiber composites have yet to be conducted. In this study, we further explore dual component polyolefin nonwoven fabrics derived from LDPE/PP and HDPE/PP co-extruded fiber composites by additional consolidation techniques. The co-extruded fiber composites can be effectively drawn in a uniaxial direction prior to delamination, thereby tuning mechanical robustness, pore size and permeability of the final nonwoven fabric. The inclusion of poly-methyl methacrylate (PMMA) colloidal nanoparticles is also explored as a potential avenue to further decrease and control the pore size and

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<https://doi.org/10.1016/j.compscitech.2018.07.022>

Received 14 February 2018; Received in revised form 25 June 2018; Accepted 13 July 2018

Available online 20 July 2018

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permeability of the non-woven mats for battery separators.

## 2. Experimental

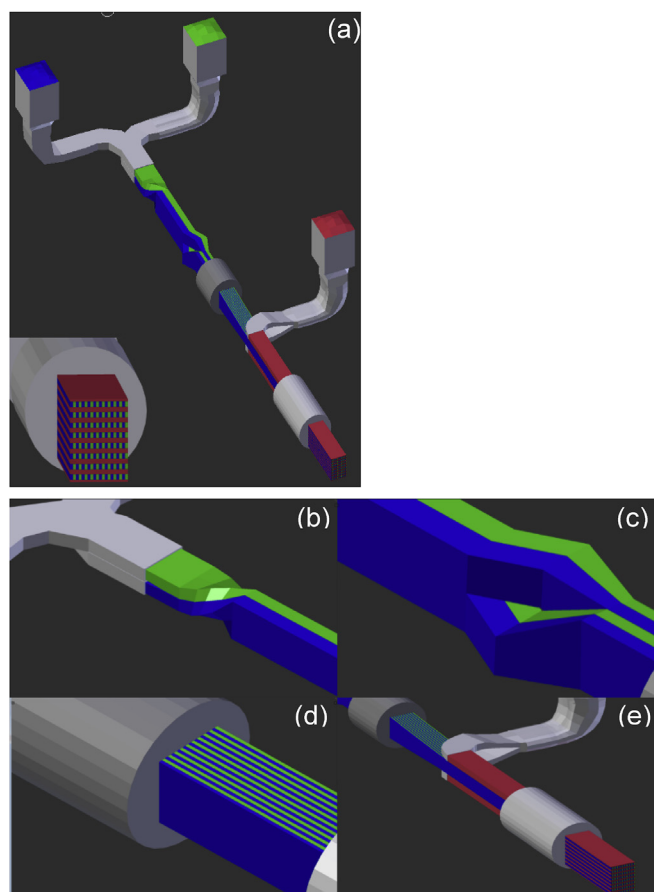
### 2.1. Materials

All chemicals were of reagent-grade and used without further purification. LDPE (Dow Chem. 5011,  $d = 0.92$ , USA), HDPE (Dow Chem. Elite G5960,  $d = 0.96$ ), PP (Exxon Mobil 2252 E4,  $d = 0.90$ , USA) and polystyrene (PS, Dow Chem. 685D  $d = 1.04$ ) were dried at 80 °C for 12 h prior to coextrusion. Potassium peroxydisulfate (KPS, CAS:7727-21-1, 99%, Sigma-Aldrich, USA), methyl methacrylate (MMA, CAS: 80-62-6, 99%, Sigma-Aldrich) and deionized water were utilized for the synthesis of poly (methyl methacrylate) (PMMA) colloidal nanoparticles. Chloroform (CAS: 67-66-3, 99%) and 1.0 M tetraethylammonium tetrafluoroborate (TEABF<sub>4</sub>, CAS:429-06-1, 99%) in propylene carbonate were supplied by Sigma-Aldrich.

### 2.2. Fabrication of fiber composite tape, fiber, and novel nonwoven fiber mat

#### 2.2.1. Fiber composite manufacturing

The novel fiber composite fabrication process (Scheme 1) is derived from traditional multilayer co-extrusion for the production of



**Scheme 1.** a: Overall scheme of coextrusion for extruded fiber composites (the inset shows the structure of final coextruded composite), b: vertical rotation of melt flow by the first vertical layer multiplier, c: generation of four vertical layers by the second vertical layer multiplier, d: generation of  $2^n$  vertical layers by ' $n$ ' vertical layer multipliers, e: introduction of sacrificing layer onto the top and the bottom of the main flow and vertical stacking by ' $m$ ' horizontal layer multipliers, producing continuous aligned domains with  $2^{n-m}$  vertical columns and  $2^m$  horizontal rows (inset of a). Green and blue colors indicate pre-fiber materials whereas red color shows a sacrificing layer.

horizontal, gradient, and vertical layered structures with individual layer thickness reaching to nanoscale [16,19]. Multilayer co-extrusion melts and combines two immiscible polymers into a stacked, single flow. The melt fluid is vertically rotated (Scheme 1b). The flow passes through a series of " $n$ " vertical layer multipliers, which split, spread, and recombine the flow, thereby producing a system of  $2^n$  vertical layers (Schemes 1c, d). A sacrificial skin layer (PS) is extruded above and below the vertical layer section (Scheme 1e). The flow then passes through a series of " $m$ " horizontal layer multipliers to produce a composite tape containing continuous aligned domains with  $2^{n-m}$  vertical columns and  $2^m$  horizontal rows (inset of Scheme 1a).

To facilitate delamination of the fiber precursor composites using mechanical separation, poor interfacial adhesion is preferable. Interphase thickness should be less than 3 nm to induce poor interfacial adhesion according to theories by Helfand, Tagami, Hansen and Hildebrand [20–22]. Coextrusion temperature was chosen for each system to achieve a viscosity match among all three materials (Fig. S1). Viscosity was determined using a melt flow index (MFI, Galaxy I Model D7054 Kayeness Inc. NJ, USA) at a shear rate comparable to extrusion conditions ( $10\text{ s}^{-1}$ ). Each composite system was coextruded through a series of 15 vertical multipliers and 6 horizontal multipliers, thereby producing 512 by 64 PE/PP layers. PS was utilized as a separating and sacrificing material since glassy PS exhibits extremely low adhesion to PE and PP [16]. Melt pump rate between the PS sacrificial layer and alternating PE/PP polyolefin domains was chosen to set the volume composition of 9:91. Melt pump rate between PE and PP was selected to set 70:30, 50:50, and 30:70 (three polyolefin compositions) for each system (LDPE/PP/PS and HDPE/PP/PS). For instance, HDPE50/PP50/PS denotes a composite containing 45.5 vol% HDPE, 45.5 vol% PP, and 9.0 vol% PS. The system containing LDPE was coextruded at 230 °C and the other containing HDPE was coextruded at 250 °C.

#### 2.2.2. Drawing

Composites (fiber precursors) were drawn at 500%/min at 105 °C with various draw ratios of 2–11, using a Zwick/Roell mechanical tester with a gauge length of 10 mm. A temperature slightly above the glass transition temperature (DSC heating at 10 °C/min) of PS (102 °C), but below the melting points of LDPE (110 °C) and HDPE (135 °C) was selected as orientation temperature. Thus, the PS domains were softly drawn, maintaining the mechanical integrity of PE/PP domains. The drawn composites were cooled down to 25 °C under tension and ambient atmosphere.

#### 2.2.3. Delamination

The composites were delaminated by using a custom water jet with a nozzle diameter of 0.01" (1600 psi, Atomizing Systems, NJ, USA). In addition to PS removal, the delamination process separates the PE domains from the PP domains, leaving a dual component rectangular PE/PP fiber system (see Scheme 2).

#### 2.2.4. Consolidation

In addition to the limited consolidation achieved through hydro-entanglement during delamination, non-woven fabric mats were further consolidated using a combination of needle punching [23] (Scheme 3) and compression molding at 200 psi for 5 min at 80 °C and 105 °C for LDPE/PP and HDPE/PP systems, respectively. The needle punching process is illustrated in Scheme 3. Manually needle-punched nonwoven mats can be fabricated by interlocking the fibers. This mechanical interlocking was performed with barbed needles repeatedly passed into and out of the mat. As the needle moved up and down, the blades of the needles penetrated the fiber bundle. Barbs on the blade of the needle picked up fibers on the downward movement and carried the fibers in the penetration direction. As the needles penetrated the fiber bundle, the fibers became increasingly entangled and interlocked. Subsequently, the needle-punched mats were softly compressed at 80 °C for 1 min under 200 psi to make the mats dense and strong.

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