Foam/Film Alternating Multilayer Structure with High Toughness and Low Thermal Conductivity Prepared via Microlayer Coextrusion

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Abstract Multilayer membranes prepared via microlayer coextrusion have attracted wide attention due to their unique properties and broad applications. In present study, the foam/film alternating multilayer sheets based on ethylene-vinyl acetate copolymer (EVA) and high-density polyethylene are successfully prepared via microlayer coextrusion. The cells in the sheets are single-cell-array along the foamed EVA layers with uniform cell size. In addition, the effects of layer number and foam relative thickness on morphology, mechanical properties, damping and heat insulation properties are investigated. The cell size decreases significantly with increasing layer number due to the enhanced confine effects. The tensile strength, elongation at break, and heat insulation also increase significantly. However, the mechanical damping properties change little in the observed frequency. Meanwhile, with higher relative thickness of EVA foam, the sheets have lower tensile strength and lower thermal conductivity, while the damping properties are enhanced in a specific frequency scope. The elongation at break of the optimized sample comes to 800% and the thermal conductivity decreases to 61 mW·m\textsuperscript{-1}·K\textsuperscript{-1}, which shows high toughness and low thermal conductivity, indicating a possible method for preparing materials with high toughness and heat-insulating properties.

Keywords Multilayer; Foam; Coextrusion; Toughness; Thermal conductivity

INTRODUCTION

Polymeric foams are widely used in packaging, insulation, aeronautic, automotive, and construction industries due to their light weight, high specific strength, low thermal conductivity, and high performance cushion compared to non-foamed polymers.\textsuperscript{[1−6]} However, the conventional bulky foams for thermal insulation usually have poor mechanical properties, which are seldom used alone. In daily applications, polymeric foams often need to be glued or bonded with other high strength materials in order to distribute load over broad areas. The process is time-consuming, complex and costly.\textsuperscript{[7]} A technique based on the coextrusion method to produce foam-sandwiched structural composite with polymeric foams as core layer and solid plastic as skin layers was proposed, which shows higher strength, better tear resistance, and improved impact resistance. However, the foam-sandwiched structure obtained by this technique only contains single or several foam layers due to the limited number of extruders.\textsuperscript{[8,9]}

Microlayer coextrusion refers to two or more polymer melts extruded in dies to form multiple layers by superimposing, splitting, and converging. The number of layers can reach as high as thousands with two or three extruders,\textsuperscript{[10,11]} and the thickness of a single layer can reach even nanoscale close to the lamella thickness of polymeric crystal.\textsuperscript{[12−14]} This technique allows to combine the desirable properties of different polymers into one structure, which has been widely used to produce high barrier packaging, electromagnetic shielding, high-efficiency sound absorbing, high-capacity information storage media, advanced capacitors, biomedical scaffolds, microporous filtration, and lithium-ion battery separators.\textsuperscript{[15−17]} When the thickness of the single layer comes to nanoscale, the nano-confine effect would be generated, and the molecular chains and crystals are organized differently from normal state.\textsuperscript{[18,19]} In terms of performance, the multilayer material shows completely different properties in the fields of mechanical,\textsuperscript{[20,21]} optical,\textsuperscript{[22,23]} insulation,\textsuperscript{[24−26]} and electromagnetic properties.\textsuperscript{[27,28]}

Microlayer coextrusion has also been employed to prepare multilayer alternating foamed structure,\textsuperscript{[29−31]} Baer et al. firstly reported multilayer foam/film structure by microlayer coextrusion,\textsuperscript{[29]} The structure consisted of alternating soft foam
layers and solid film layers. The comprehensive behavior of 8–64 layers polypropylene (PP) foam/PP film structures was similar to that of cork; the tensile and compressive properties of the material increased with increasing number of layers, which greatly improved the mechanical properties of the foamed materials. The optimal sample broke at 8 MPa and 12% strain. Rahman et al. developed low-density polyethylene (LDPE) multilayer foam/film systems with viscosity contrast between film and foam layer polymers. They found high viscosity contrast contributed to layer integrity and intact closed-cells. With higher layer number, the confine effect on cell growth increased significantly, resulting in single cell alignment in the foam layers. Sun et al. used linear low-density polyethylene (LLDPE)/poly(ethylene-octene) elastomer (POE) blend as the film layers and cross-linked POE as the foam layers. They found the sound absorption coefficient of multilayer foam/film structure was 2–3 times higher than that of conventional single-layer material in selected frequency range. Moreover, they optimized single layer thickness and ratio of foam/film layers to obtain the most efficient sound absorption at specific frequency.

As mentioned before, most work on foam/film multilayer structure in recent years mainly focused on the acoustic and mechanical properties of such structure, while other aspects of physics properties were rarely paid attention. In the present study, effects of layer number and foam relative thickness on the heat-insulating and mechanical damping properties of foam/film alternating multilayer structure are investigated. In addition, major literatures chose polyolefin as foam material, which limits property exploration and potential applications. How to control the foam stability in polyolefin melt is the most important. Other than the method of cross-linking, using the higher viscosity melt as the confining film layer and lower viscosity melt as the foam layer in multilayer coextrusion may become a useful method for preparing alternately multilayered foam/film structure. In this work, ethylene-vinyl acetate copolymer (EVA)/high-density polyethylene (HDPE) foam/film alternating multilayer structure was prepared via microlayer coextrusion, which may greatly improve the elongation at break of the material, and overcome the disadvantage of easy breaking of the conventional foam structure, providing a new direction to prepare materials with high toughness and low thermal conductivity.

**EXPERIMENTAL**

**Materials**

High-density polyethylene (HD3840UA, melt index (MI)=4.0 g·10min⁻¹) was obtained from Shanghai Gaoqiao Petrochem. Co. Ethylene-vinyl acetate copolymer (7350M, MI=2.5 g·10min⁻¹) was bought from Shanghai Gaoqiao Petrochem. Co. CaCO₃ powder with an average particle size of 800 nm was purchased from Aladdin Chem. Co.

**Preparation of EVA Foam/HDPE Film Alternating Multilayer Structure**

EVA foam/HDPE film alternating multilayer structure was prepared on a home-made microlayer coextrusion device. The previously prepared master batch of EVA, nucleating agent (CaCO₃), and chemical blowing agent (azodicarbonamide) was fed into one extruder, while HDPE was fed into the other extruder. These two melt streams were merged in the feed block to form two parallel layers, and then multilayer structure was achieved after flowing through the section of layer multipliers. During extrusion, the temperature of the extruders and multiplier elements was set at 180 °C, and that of the foam die was set at 200 °C. The screw rotating rate of the extruder for HDPE was fixed at 10 r·min⁻¹, while that of the extruder for EVA was decided by volume ratio of foam/film. The schematic diagram of the preparation for foam/film alternating multilayer material is shown in Fig. 1. The formulations with different layer numbers and volume ratios of foam/film are listed in Table 1. For comparison, the EVA/HDPE blends were prepared in the mixer. The neat EVA, neat HDPE, solid blend sheets, and blend foams were prepared on the hot press. The proportion of EVA/HDPE blends and the blend foams is shown in Table 2.

**Table 1** Overview of EVA foam/HDPE film alternating multilayer sheets via microlayer coextrusion taking 8 layers as example.

| Sample | Layers | AC content (wt%) | V_foam/V_film |
|--------|--------|------------------|---------------|
| 8L, 0/1 | 8      | 0.5              | 1:1           |
| 8L, 1/1 | 8      | 0.8              | 1:1           |
| 8L, 1/0, 0.8% | 8 | 1                | 1:1           |
| 8L, 1/1, 1% | 8 | 0.8             | 1:1           |
| 16L, 1/1 | 16     | 0.8              | 1:1           |
| 32L, 1/1 | 32     | 0.8              | 1:1           |
| 32L, 2/1 | 32     | 0.8              | 2:1           |
| 32L, 3/1 | 32     | 0.8              | 3:1           |

**Table 2** Proportion of EVA/HDPE blends and blend foams.

| Sample | Layers | AC content (wt%) | V_foam/V_film | Temperature (°C) |
|--------|--------|------------------|---------------|-----------------|
| 1L, 0/1 | 1      | 0                | 0.1           | 200             |
| 1L, 1/0 | 1      | 0.8              | 1:1           | 200             |
| EVA    | 1      | 0                | 0.1           | 200             |
| HDPE   | 1      | 0                | 0.1           | 200             |

**Characterization**

The foam/film layered structure in each sample was observed by optical microscopy (KEL-XMT-3100 from Nanjing Kyle Instrument Co.). The samples were cut into approximately 50 μm thickness by sharp blades perpendicular to extrusion direction at room temperature. The cell distribution and the layer stratification in the samples were observed by transmission mode. At the same time, more than 30 cells were counted by
Nano Measurer 1.2 software to calculate the average diameter of the cells in the foams.

The tensile properties in extrusion direction of the samples were measured according to GB/T 1040.3-2006 on universal mechanical testing machine (UTM6501, SANS Test Mechatanical Machines Co.) and fixture (JDSAS20A, SANS Test Mechatanical Machines Co.) with a strain rate of 10 mm·min⁻¹. Interlayer peel strength of multilayer unfoamed samples was tested according to GB/T 8808-1998 on universal mechanical testing machine (UTM6501, SANS Test Mechanical Machines Co.) and fixture (JDSAS20A, SANS Test Mechanical Machines Co.). The compressive properties perpendicular to the extrusion direction of the samples were measured according to GB/T 20467-2006 on universal mechanical testing machine (UTM6501, SANS Test Mechanical Machines Co.) and load cell (JYA204A, SANS Test Mechanical Machines Co.) with a strain rate of 0.25 mm·min⁻¹.

Damping properties of the samples were evaluated using dynamic thermomechanical analysis (Q800, TA Instruments). Rectangle samples of 60 mm × 10 mm × 1.2 mm were used in all tests. Measuring condition was set to dual cantilever mode, amplitude was 25 µm, and the frequency was set to 1−50 Hz. All tests were performed at 30 °C. Loss tangent (tanδ) is used to measure the energy loss during the vibration.

Thermal conductivity of the samples was measured on a Lambda heat flow thermal conductivity analyzer (HFM446, Netzsch Co.) under steady-state heat flow condition. The upper plate temperature was set at 10 °C as the hot plate and the lower plate temperature was set at −10 °C as the cold plate, with a temperature difference of 20 °C. All tests were repeated three times to get the average value. The thermal conductivity (λ) can be calculated using Eq. (1) as follows:

\[ \lambda = NL \left( \frac{q_T}{\Delta T_m} \right) \]  

(1)

where \( N \) is a calibration factor, \( L \) is the sample thickness, \( q_T \) is the heat flux and \( \Delta T_m \) is temperature difference.

To measure the apparent density (\( \rho \)) of the samples, three specimens for each sample were cut in width of 120 mm and length of 120 mm. The weighting method was used. All tests were repeated three times to get the average value. The apparent density can be calculated by Eq. (2) as follows:

\[ \rho = \frac{m}{V} \]  

(2)

where \( m \) is the mass of the sample and \( V \) is the volume.

RESULTS AND DISCUSSION

Preparation and Morphology of EVA Foam/HDPE Film Alternating Multilayer Sheets

EVA foam/HDPE film alternating multilayer materials were prepared via microlayer coextrusion, as shown in Fig. 1. EVA, nucleating agent (CaCO₃), and chemical blowing agent were previously prepared into the master batch, which was then fed into one extruder, while HDPE was fed into the other one. The multilayer structure was obtained after flowing through the section of layer multipliers, in which the blowing agent started to decompose and produce a large amount of gas to develop foam layers at the same time. Effects of the content of AC blowing agent on morphology of foam/film alternating multilayer materials are shown in Fig. 2. The dark regions are EVA foam layers and the bright parts are HDPE film layers. In EVA/HDPE multilayer system, adjacent layers have obvious delamination and good interlayer adhesion; the peel strength between layers of sample 8L, 0/1 is shown in Fig. S1 (in the electronic supplementary information, ESI). From Figs. 2(a)–2(c), the cells are distributed in single-cell-array along the EVA layers. The surface EVA layer has no cell distribution due to the fact that there is only one single HDPE film layer adjacent to the surface EVA layer, which cannot cause confine effect to limit cell growth and prevent gas escaping. With AC blowing agent increasing from 0.5 wt% to 1 wt%, the number and size of cells increase. When blowing agent comes to 0.8 wt%, the number and size of cells are moderate, and the multilayer structure is not damaged. Therefore, the content of blowing agent was fixed at 0.8 wt% in the following foaming samples. Fig. 3 shows the cross-section of EVA foam/HDPE film alternating multilayer sheets with different foam thicknesses. The foam/film alternating multilayer sheets exhibit a completely different multilayer structure from EVA/HDPE blend foam or unfoamed EVA/HDPE multilayer sheet. The relative thickness of EVA foam also has a significant influence on the size of the foam cells.

Fig. 4 is the cell diameters of different samples counted from Figs. 2 and 3. As shown in Fig. 4, with the thickness of single layer decreasing, the average diameter of the cell decreases significantly from 235 µm at 8 layers to 89 µm at 32 layers. The reason is that the closed environment between the film layers has a strong confine effect on the cell growth. The higher the layer number is, the more enhanced the confine effect will be. The cell diameter decreases obviously. At the same time, the cell diameter increases from 89 µm at \( V_{\text{foam}}:V_{\text{film}}\) 1:1 condition to 120 µm at \( V_{\text{foam}}:V_{\text{film}}\) 3:1 condi-

![Fig. 2](https://doi.org/10.1007/s10118-021-2524-0)
tion. More gas generated by the decomposition of the blowing agent and weaker inter-layer confine effect of the films both lead the cells to grow more easily, which contributes to larger cell diameter.

Tensile and Compressive Properties of EVA Foam/HDPE Film Alternating Multilayer Sheets

Fig. 5 displays the tensile properties of EVA foam/HDPE film alternating multilayer sheets with different layer numbers and foam relative thicknesses. The tensile strength and elongation at break of the samples are shown in Table 3. In EVA foam/HDPE film alternating multilayer sheets, solid HDPE films play a major role in mechanical bearing. Therefore, the tensile behavior of the system as a whole is mainly contributed by the tensile properties of solid HDPE. In each stress-strain curve, the strain initially shows a short linear elastic deformation stage and then gradually increases as the stretch increases due to work hardening effect. In the stretching process, the cells would bend and stretch, and the densification would appear after cells collapse. Compared with 8 layers unfoamed sample, tensile strength and elongation at break of 8 layers foamed sample reduce significantly. With the increase of multilayer number from 8 layers to 32 layers, the elongation at break improves from 600% to 800% and the tensile strength at break increases

Table 3 Tensile properties of EVA foam/HDPE film alternating multilayer sheets.

| Sample | Tensile strength (MPa) | Elongation at break (%) |
|--------|------------------------|-------------------------|
| 8L, 0/1| 14.83±1.24             | 956±74                  |
| 8L, 1/1| 6.18±0.52              | 648±51                  |
| 16L, 1/1| 7.85±0.57             | 703±47                  |
| 32L, 1/1| 10.41±0.89             | 855±81                  |
| 32L, 2/1| 10.22±0.74             | 842±72                  |
| 32L, 3/1| 10.13±0.91             | 849±68                  |

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from 6.5 MPa to 10 MPa. With increasing number of multilayers, the orientation of the molecular chain along the extrusion direction is enhanced in the extrusion process.\textsuperscript{[13]} Moreover, more layers would cause smaller cells in the material, which means fewer and smaller defects. It is harder to break when stretched. With higher EVA foam content in the samples, the yielding stress decreases, while elongation at break is almost unchanged. It should be attributed to the reason that the mechanical properties of the multilayer structure along the extrusion direction are mainly provided by the HDPE layer, so the tensile strength decreases on account of less HDPE film content but the elongation at break keeps constant.

The compressive stress-strain curves of various samples are displayed in Fig. 6. In each compressive stress-strain curve, it initially shows a short linear region and then a long plateau; finally the stress rises rapidly as the stretch goes. In the initial stages of the compression, the cells would bend and deform, reflecting linear elastic modulus. When cells begin to collapse, a long plateau would show in the stress-strain curve. After cells collapse, the densification would appear and the curve would rise sharply. As seen in Fig. 6(a), the compressive modulus and densification strain increase with increasing layer number. Fig. 6(b) shows higher foam content would cause lower compressive modulus but higher densification strain.

**Dynamic Thermomechanical Analysis of EVA Foam/HDPE Film Alternating Multilayer Sheets**

The storage modulus represents the ability to store elastic deformation energy and is an index to measure rigidity. Fig. 7 displays dynamic mechanical analysis of EVA foam/HDPE film alternating multilayer sheets. As shown in Fig. 7(a), compared with 8 layer unfoamed EVA/HDPE alternating multilayer sheets, all multilayer foamed samples have much lower storage modulus in the frequency range of 1−50 Hz. As the layer number increases and EVA foam relative thickness decreases, the storage modulus increases. The reason is that higher layer number would lead to enhanced polymer chain orientation, which means higher rigidity and storage modulus. Thinner foam layer would also cause higher strength and storage modules. The results are consistent with the stretch and compression results above.

![Fig. 6](image1)

**Fig. 6** (a) Effects of layer number on compressive behavior of EVA foam/HDPE film alternating multilayer sheets; (b) Effects of EVA foam thickness on compressive behavior of foam/film alternating multilayer sheets.

![Fig. 7](image2)

**Fig. 7** (a) Storage modulus and (b) tanδ of EVA foam/HDPE film alternating multilayer sheet with different layer numbers and foam thicknesses.

The ratio between the storage modulus and the loss modulus is tanδ, which can be used to measure the energy loss during vibration. Fig. 7(b) shows that all foam/film alternating multilayer sheets have higher tanδ than 8 layer unfoamed EVA/HDPE alternating multilayer sheets. The reason is that the energy dissipation mainly depends on the friction between the polymer segments when an unfoamed sample is deformed by external forces. The alternating multilayer foam-
ing structure introduces a large number of cells. When the structure is deformed, apart from polymer chain friction, the deformation and recovery of the cells will transform a large amount of energy into heat energy, resulting in higher loss tangent.\[33\] Theoretically, differences of interface properties during the vibration will also cause more energy consumption with increasing layer number.\[34\] However, the difference of tanδ between 8 and 32 layers is not significant in this work. The reason may be that EVA and HDPE layers have good interfacial adherence, resulting in little energy loss caused by layer interface during vibration; hence, the effects of layer number on tanδ is not obvious in EVA/HDPE system.

**Effects of Layer Number and Foam Relative Thickness on Thermal Conductivity**

Thermal conductivity is an important index to measure the thermal insulation capability of foamed materials. As shown in Fig. 8(a), the layer number and the foam relative thickness have great impacts on thermal conductivity of the samples. The thermal conductivity of neat EVA, neat HDPE, and common EVA/HDPE blends are 111.4, 105.8 and 107 mW·m\(^{-1}\)·K\(^{-1}\), respectively. When the blends are foamed, the thermal conductivity reduces to 89 mW·m\(^{-1}\)·K\(^{-1}\). With the layer number increasing from 8 to 32, the thermal conductivity of EVA foam/HDPE film alternating multilayer sheets decreases from 84 mW·m\(^{-1}\)·K\(^{-1}\) to 71 mW·m\(^{-1}\)·K\(^{-1}\). Higher foam relative thickness in multilayer structure would also cause lower thermal conductivity. The minimum thermal conductivity is 61 mW·m\(^{-1}\)·K\(^{-1}\) at 32 layers and \(V_{\text{foam}}:V_{\text{film}}=3:1\) condition, which is only 57% of that of common EVA/HDPE blends.

The results can be explained by heat transfer mechanism, which includes three methods (heat conduction, heat radiation, and heat convection). When the environment temperature is lower than 200 °C and cell size in porous material is less than 4 mm, heat radiation and heat convection could be ignored, and heat conduction becomes the major heat transfer way.\[35\] The heat conduction in foamed materials is composed of gas and polymer heat conduction, and the thermal conductivity of gas is much lower than that of the polymer. It can explain why the samples with higher foam relative thickness show lower thermal conductivity.\[36\] When layer number increases, the sample density does not change greatly as displayed in Fig. 8(b), while the thermal conductivity decreases significantly. The reason is that the heat energy in polymer is mainly transmitted through the irregular diffusion of phonons at the interface of EVA foam and HDPE film layer. The scattering of phonons is intensified, which impedes the transmission of phonons and effectively reduces the thermal conductivity.\[37,38\]

**CONCLUSIONS**

EVA foam/HDPE film alternating multilayer materials are successfully prepared via microlayer coextrusion. The cells with uniform size in the materials show single-cell-arrangement along the EVA foaming layers. The cell diameter decreases from 245 μm at 8 layers to 89 μm at 32 layers due to significant confine effect. The tensile strength and the heat insulation increase with increasing layer number, while the damping properties change little. Higher foam relative thickness would cause weaker tensile strength, lower thermal conductivity, and higher damping properties at specific frequency range. However, the elongation at break basically keeps constant. After optimization, the elongation at break reaches up to 800% with the thermal conductivity as low as 60 mW·m\(^{-1}\)·K\(^{-1}\), showing high toughness and good heat insulation.

**Electronic Supplementary Information**

Electronic supplementary information (ESI) is available free of charge in the online version of this article at https://doi.org/10.1007/s10118-021-2524-0.

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REFERENCES

1. Suh, K. W.; Park, C. P.; Maurer, M. J.; Tusin, M. H.; Genova, D. R.; Broos, R.; Sophiea, D. P. Lightweight cellular plastics. *Adv. Mater.* **2000**, *12*, 1779–1789.

2. Rodriguez-Perez, M. A. Crosslinked polylefin foams: production, structure, properties, and applications. *Adv. Polym. Sci.* **2005**, *184*, 97–126.

3. Wang, J.; Zhang, L.; Bao, J. B. Supercritical CO2 assisted preparation of open-cell foams of linear low-density polyethylene and linear low-density polyethylene/carbon nanotube composites. *Chin. J. Polym. Sci.* **2016**, *34*, 889–900.

4. Sorrentino, L.; Aurilia, M.; Iannace, S. Polymeric foams from high-Performance thermoplastics. *Adv. Polym. Technol.* **2011**, *30*, 234–243.

5. Zhang, R. Z.; Chen, J.; Huang, M. W.; Zhang, J.; Luo, Q. G.; Wang, B. Z.; Li, M. F.; Shen, Q.; Zhang, L. M. Synthesis and compressive response of microcellular foams fabricated from thermally expandable microspheres. *Chin. J. Polym. Sci.* **2019**, *37*, 279–288.

6. Zia, K. M.; Bhatti, H. N.; Bhatti, I. A. Methods for polyurethane and polyurethane composites, recycling and recovery: a review. *React. Funct. Polym.* **2017**, *67*, 675−692.

7. Shetty, R.; Han, C. D. Study of sandwich foam coextrusion. *J. Appl. Polym. Sci.* **1978**, *22*, 2573–2584.

8. Passaro, A.; Corvaglia, P.; Manni, O.; Barone, L.; Maffeizoli, A. Processing-properties relationship of sandwich panels with polypropylene-core and polypropylene-matrix composite skins. *Polym. Compos.* **2004**, *25*, 307−318.

9. Dukess, J. *1979*, US4107247-A.

10. Schrenk, W. J.; Bradley, N. L.; Alfrey, T.; Maack, H. Interfacial flow instability in multilayer coextrusion. *Polym. Eng. Sci.* **1978**, *18*, 620–623.

11. Schrenk, W. J.; Wheatley, J. A.; Lewis, R. A.; Arends, C. B. Nanolayer polymeric optical films. *Tappi J.* **1992**, *75*, 169–174.

12. Carr, J. M.; Langhe, D. S.; Ponting, M. T.; Hiltner, A.; Baer, E. Confined crystallization in polymer nanolayered films: a review. *J. Mater. Res.* **2012**, *27*, 1326–1350.

13. Cheng, J. F.; Schrenk, W. J.; Liu, H. T. Orientation of LDPE crystals from microscale to nanoscale via microlayer or nanolayer coextrusion. *Chin. J. Polym. Sci.* **2016**, *34*, 1411–1422.

14. Rahman, M. A.; Zhang, J.; Zhang, C.; Olah, A.; Baer, E. Novel micro-/nano-porous cellular membranes by forced assembly coextrusion technology. *Eur. Polym. J.* **2016**, *83*, 99–113.

15. Cheng, J. F.; You, X. H.; Cao, Z.; Wu, D.; Liu, C. L.; Pu, H. T. Effective control of laser-induced carbonization using low-density polyethylene/polystyrene multilayered structure via nanolayer coextrusion. *Mater. Sci. Eng. C* **2019**, *101*, 1800726.

16. Li, Z. P.; Olah, A.; Baer, E. Micro- and nano-layered processing of new polymeric systems. *Prog. Polym. Sci.* **2020**, *102*, 101210.

17. Li, Y. J.; Pu, H. T. Facile fabrication of multilayer separators for lithium-ion battery via multilayer coextrusion and thermal induced phase separation. *J. Power Sources* **2018**, *384*, 408–416.

18. Tan, X.; Li, J.; Guo, S. Y. Temperature-dependent order-to-order transition of polystyrene-block-poly(ethylene-co-butylene)-block-polystyrene triblock copolymer under multilayered confinement. *Macromolecules* **2018**, *51*, 2099–2109.

19. He, Y. N.; Liu, X.; Liao, Q.; Zhou, J. J.; Huo, H.; Li, L. Thickness-dependent orientation structure in poly(ethylene oxide) multilayer crystals. *Chin. J. Polym. Sci.* **2014**, *32*, 1253–1259.

20. Li, C. H.; Wang, J. F.; Guo, J. W.; Wu, Hong.; Guo, S. Y. The toughening behavior of the PP/POE alternating multilayered blends under EWF and impact tensile methods. *Chin. J. Polym. Sci.* **2015**, *33*, 1477–1490.

21. Montana, J. S.; Roland, S.; Richaud, E.; Miquelard, G. G. From equilibrium lamellae to out-of-equilibrium cylinders in triblock copolymer nanolayers obtained via multilayer coextrusion. *Polymer* **2018**, *136*, 27–36.

22. Li, Z. P.; Sun, R. L.; Rahman, M. A.; Feng, J. X.; Olah, A.; Baer, E. Scaling effects on the optical properties of patterned nanolayered shape memory films. *Polymer* **2019**, *167*, 182–192.

23. Ryan, C.; Christenson, C. W.; Vallee, B.; Saini, A.; Lott, J.; Johnson, J.; Schiraldi, D.; Weder, C.; Baer, E.; Singer, K. D.; Shan, J. Roll-to-roll fabrication of multilayer films for high capacity optical data storage. *Adv. Mater.* **2012**, *24*, 5222–5226.

24. Wang, H. P.; Keum, J. K.; Hiltner, A.; Baer, E.; Freeman, B.; Rozanski, A.; Galeski, A. Confined crystallization of polyethylene oxide in nanolayer assemblies. *Science* **2009**, *323*, 757–760.

25. Messin, T.; Follain, N.; Guinaud, S.; Sollogoub, C.; Gaucher, V.; Delpouve, N.; Marais, S. Structure and barrier properties of multilayered biodegradable PLA/PBSA films: confinement effect via forced assembly coextrusion. *ACS Appl. Mater. Interfaces* **2017**, *9*, 29101–29112.

26. Messin, T.; Marais, S.; Follain, N.; Chappey, C.; Guinaud, A.; Miquelard-Garnier, G.; Delpouve, N.; Gaucher, V.; Sollogoub, C. Impact of of water and thermal induced crystallizations in a PC/MXD6 multilayer film on barrier properties. *Eur. Polym. J.* **2019**, *111*, 152–160.

27. Mackey, M.; Schuele, D. E.; Zhu, L.; Flandon, L.; Wola, M. A.; Shirk, J. S.; Hiltner, A.; Baer, E. Reduction of dielectric hysteresis in multilayer films via nanocoordination. *Macromolecules* **2012**, *45*, 1954–1962.

28. Ji, X. Y.; Chen, D. Y.; Shen, J. B.; Guo, S. Y. Flexible and flame-retarding thermoplastic polyurethane-based electromagnetic interference shielding composites. *Chem. Eng. J.* **2019**, *370*, 1341–1349.

29. Ranade, A. P.; Hiltner, A.; Baer, E.; Bland, D. G. Structure-property relationships in coextruded foam/film microlayers. *J. Cell. Plast.* **2004**, *40*, 497–507.

30. Rahman, M. A.; Andrade, R.; Maia, J.; Baer, E. Viscosity contrast effects on the structure—property relationship of multilayer soft film/foams. *Polymer* **2015**, *69*, 110–122.

31. Sun, X. J.; Liang, W. B. Cellular structure control and sound absorption of polyolefin microlayer sheets. *Compos. Part B* **2016**, *87*, 21–26.

32. Li, Y. J.; Pu, H. T.; Wei, Y. L. Polypropylene/polyethylene multilayer separators with enhanced thermal stability for lithium-ion battery via multilayer coextrusion. *Electrochem. Acta* **2018**, *264*, 140–149.

33. Zhang, X. X.; Lu, Z. X.; Tian, D.; Li, H.; Lu, C. H. Mechanochemical devulcanization of ground tire rubber and its application in acoustic absorbent polyurethane foamed composites. *J. Appl. Polym. Sci.* **2013**, *127*, 4006–4014.

34. Xu, K. M.; Huo, Q. M.; Wu, H.; Guo, S. Y.; Zhang, F. S. Designing a polymer-based hybrid with simultaneously improved mechanical and damping properties via a multilayer structure construction: structure evolution and a damping mechanism. *Polymers* **2020**, *12*, 446.

35. Long, L. S.; Ye, H. The roles of thermal insulation and heat storage in the energy performance of the wall materials: a simulation study. *Sci. Rep.* **2016**, *6*, 24181.

36. Wang, Z.; Zhang, T.; Park, B. K.; Lee, W. I.; Hwang, D. J. Minimal contact formation between hollow glass microparticles toward low-density and thermally insulating composite materials. *J. Mater. Sci.* **2017**, *52*, 6726–6740.

37. Kim, G.-H.; Lee, D.; Shanker, A.; Shao, L.; Kwon, M. S.; Gidley, D.; Kim, J.; pipe, K. P. High thermal conductivity in amorphous polymer blends by engineered interchain interactions. *Nat. Mater.* **2015**, *14*, 295–300.

38. Xie, X.; Li, D. Y.; Tsai, T. H.; Liu, J.; Braun, P. V.; Cahill, D. G. Thermal conductivity, heat capacity, and elastic constants of water soluble polymers and polymer blends. *Macromolecules* **2016**, *49*, 972–978.