Electronic Supplementary Information

Fabrication of Polymer/Aligned Shish-Kebab Composite:

Microstructure and Mechanical Properties

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1) Calculation method of the content of USKM in the USKM/LLDPE composite

As the USKM/LLDPE composite was prepared by solution casting method, both the weight of USKM and USKM/LLDPE composite must be obtained through weighing and calculation. After the shear-induced crystallization of UHMWPE/xylene solution, the USKM was collected on the ferrum frame. The USKM together with the ferrum frame could be weighed and the weight was m_1. Then LLDPE/xylene solution was casted on the USKM. After the solvent was absolutely evaporated, the USKM/LLDPE casted composite as well as the ferrum

frame was weighed and the weight was m_3. Moreover, the ferrum frame should be taken out before the USKM/LLDPE casted composite was pressed using the vacuum-assisted compression molding machine. After being taken out, the ferrum frame was weighed and the weight was m_2. Therefore, the weight of the USKM could be obtained by

$$m_{USKM} = m_1 - m_2 \qquad (1)$$

where m_{USKM} represented weight of the USKM. Meanwhile, the weight of USKM/LLDPE composite was calculated by

$$m_{composite} = m_3 - m_2 \qquad (2)$$

where $m_{composite}$ represented weight of the USKM/LLDPE composite. So, the concentration of USKM in the USKM/LLDPE composite was

$$r = \frac{m_{USKM}}{m_{composite}} \times 100\% = \frac{m_1 - m_2}{m_3 - m_2} \times 100\% \qquad (3)$$

2) Preparation of rectangular specimens for tensile test

As shown in Figure S1, rectangular specimens with a length of 10 mm and width of 5 mm were cut from centre of the compression moulded samples along the alignment direction of the shish-kebabs in USKM. The tensile direction was along the alignment direction of the shish-kebabs, *i.e.*, along the length direction of the rectangular specimens.
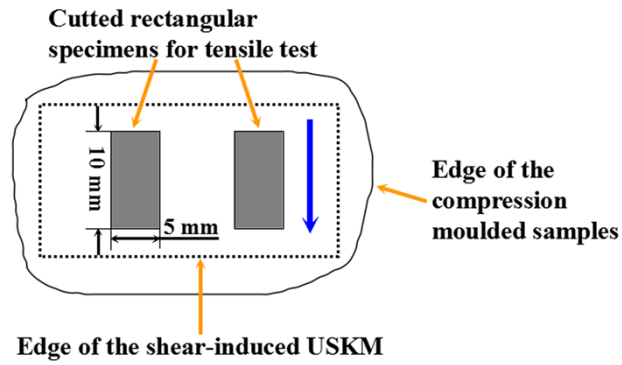
Figure S1. Schematic of the preparation of rectangular specimens for tensile test. The blue arrow refers to shear direction of the USKM.

3) Supplementary SEM images of the USKM/LLDPE composite

As shown in Figure S2, fibrils indicated by blue arrows can be clearly observed in the USKM/LLDPE composite. Owing to incomplete etching, not all fibrils can be substantially exposed. Clearly, these fibrils are embedded into LLDPE matrix.
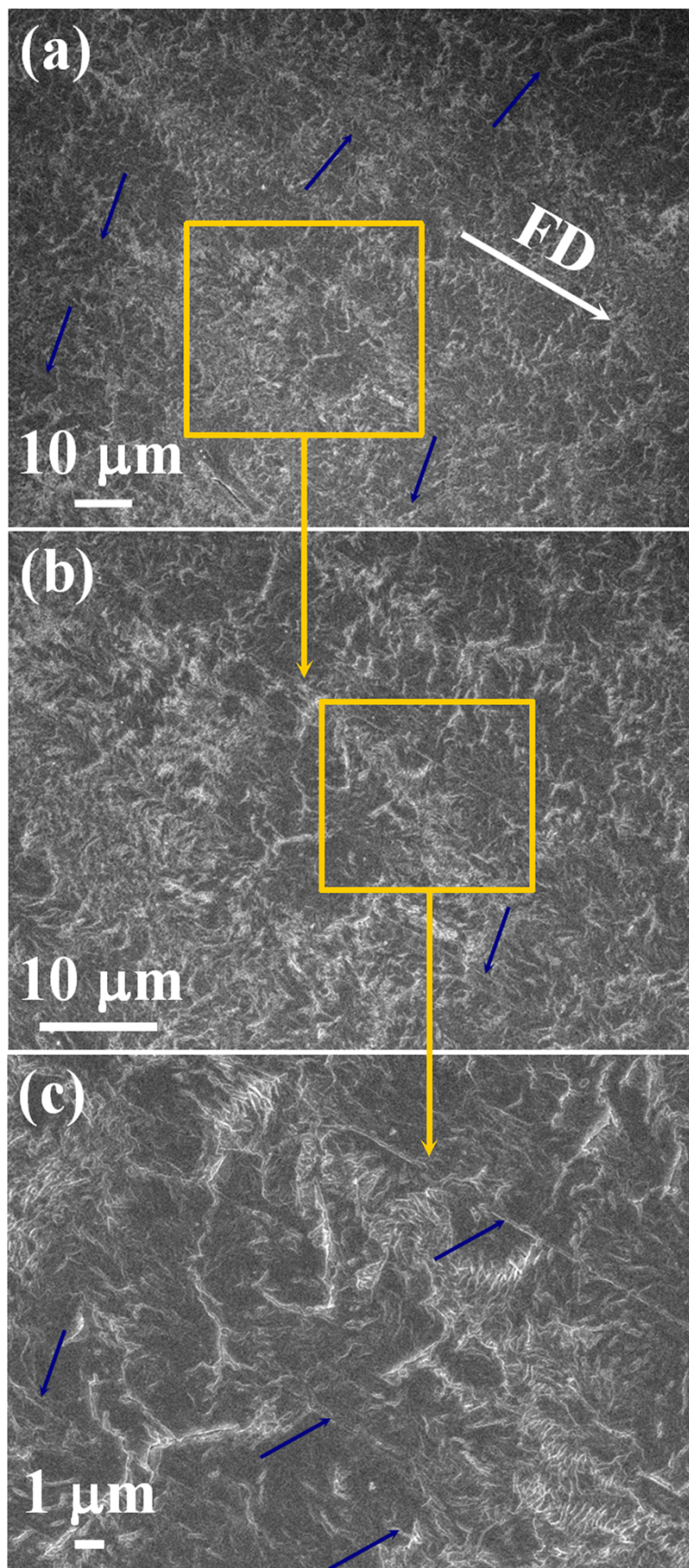
Figure S2. Supplementary SEM images of the USKM/LLDPE composite. The white arrow refers

to flow direction of shish structures.