Supplementary Information (SI) for Materials Horizons.

# Supporting Information

## Experimental section

### a. Materials and Film Preparation

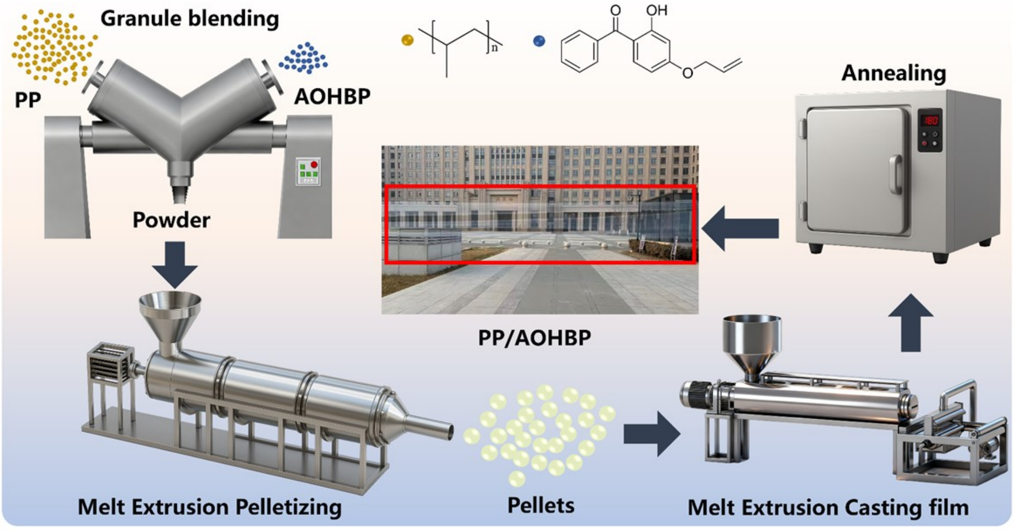


**Figure S1.** Preparation of PP/AOHBP film based on physical blending and melt extrusion casting process

Ultralow-ash PP powder was supplied by Sinopec Zhongyuan Petrochemical Co., Ltd., and industrial-grade AOHBP was used as received. To ensure smooth blending and prevent impurities from interfering with the process, the PP powder was pretreated to remove surface contaminants prior to use. The PP and AOHBP were physically compounded in different weight ratios via melt blending. Specifically, predetermined amounts of PP and AOHBP powders were first mixed in a VH-3.0 high-speed mixer to obtain PP/AOHBP powder samples with AOHBP contents of 0.4 wt%, 0.8 wt%, and 1.2 wt%, respectively. The blended powders were then fed into a twin-screw extruder (PULITAI HM50) for granulation, producing PP/AOHBP composite pellets with controlled compositions. Subsequently, the pellets were processed into thin films (~12 μm thickness) using a single-screw film-casting extruder (Leistritz ZSE 50CC). The film-casting process parameters were as follows: the melt temperature was set stepwise at 200°C, 215°C, 220°C, 225°C, and 225°C along the extruder zones; the screw rotation speed was maintained at 20 rpm, and the take-up speed of the cast film was 11 rpm. After film formation, both pure PP and PP/AOHBP films were annealed in a constant-temperature oven (Labtech Engineering Co., Ltd., LCR-300) at 140°C for 12 hours to relieve internal stresses. The resulting composite films with different AOHBP contents were labeled as PP/AOHBP-1, PP/AOHBP-2, and PP/AOHBP-3, with the suffix number increasing with higher AOHBP concentration.

### b. Instrument and characterization

The decomposition temperature ($T_{d,5\%}$) was measured using a Mettler–Toledo thermogravimetric analyzer (TGA), with samples heated from 35 °C to 800 °C at a rate of 10 K/min under a nitrogen atmosphere. The melting and crystallization temperatures were determined using a Mettler–Toledo DSC (DSC 200 Polyma), with a Netzsch aluminum crucible and an empty crucible used as reference. The crystalline structure was characterized by X-ray diffraction (XRD) using a Bruker D8 ADVANCE diffractometer. The contact angle was measured with a DSA100S goniometer. Tensile properties were evaluated using a universal testing machine. Dynamic mechanical properties were obtained using a Netzsch DMA242E dynamic mechanical analyzer (DMA). The chemical structure was analyzed by Fourier transform infrared spectroscopy (FT-IR) on a Bruker VERTEX70. Dielectric properties were measured using a Concept80 dielectric spectrometer, while energy-storage performance and leakage current were assessed using a Polyk ferroelectric tester. Breakdown strength was evaluated with a DC voltage tester. Surface morphology was examined using a GeminiSEM 500 field-emission scanning electron microscope (SEM). The electrode metal layer was deposited via magnetron sputtering using a Cicel CCZK-700SF vacuum coating system. Density functional theory (DFT) calculations were performed in Gaussian 16 using the B3LYP hybrid functional and the 6-31G basis set.

**Calculation of Crystallinity by DSC result**

The crystallinity (Xc) of PP polymer was calculated from the crystallization curves by using Equation S2,

$$X_C = \frac{\Delta H_m}{\Delta H_m^0} \times 100\% \qquad \text{(S1)}$$

Where $\Delta H_m$ is the crystallization enthalpy of PP extracted from the DSC curves and ΔHm0 is the melting enthalpy of a 100% crystalline PP sample. The value was taken as 207 J/g according to literature.

## Measurement of Dielectric Breakdown Strength

Dielectric breakdown is a statistic phenomenon, often described by the Weibull statistical model, $f(x) = 1 - \exp(-(E/\alpha)^{\beta})$, where f(x) is the probability of the films to breakdown at electric field $E$, $\alpha$ is the Weibull breakdown field at which 63.2% of the film breakdown $E_b$ and shape parameter $\beta$ is the statistical spread of the breakdown field. The breakdown strength is related to area and thickness of dielectric films, becoming lower with the test electrode area, and with increasing dielectric thickness. For example, using Weibull model, the Weibull breakdown field $\alpha$ decreases with the electrode area A, $\alpha_1 = \alpha_0 * (A_0/A_1)^{1/\beta}$.

## Calculation Method for Yield Strength.

Under the ISO 527 standard, yield strength is determined from the stress-strain curve. When a material enters the yielding phase during a tensile test, the curve exhibits significant changes, typically marked by an inflection point that indicates the onset of yielding. The specific steps for calculating yield strength are as follows:

1. Stress-Strain Curve Generation

The stress-strain data obtained from the testing machine are used to plot the curve. Ideally, the curve should display a clear elastic phase (linear stress-strain relationship) followed by the yielding phase (where stress growth becomes nonlinear, and the material undergoes plastic deformation).

2. Identification of the Yield Point

The yield point is typically defined as the point where the material transitions from linear elastic behavior to nonlinear deformation under stress.

3. The Yield strength of PP polymer was calculated from the crystallization curves by using Equation S4,

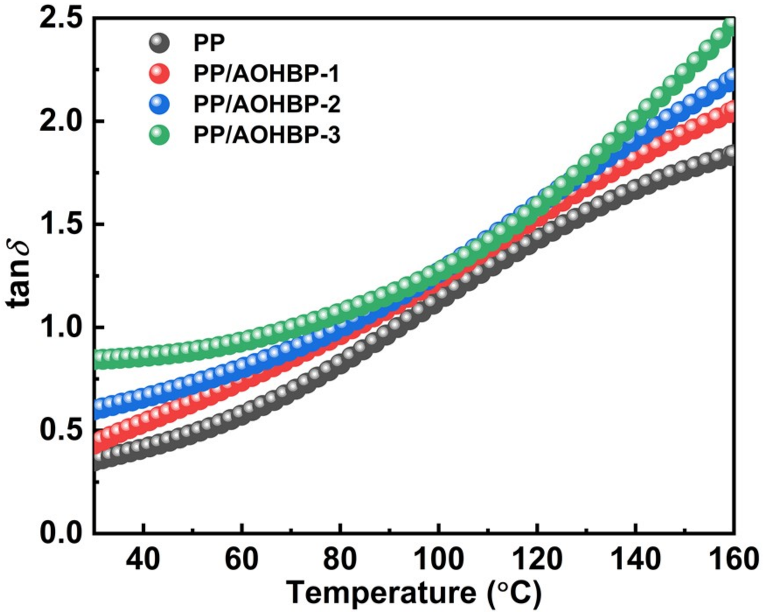
$$\delta_y = \frac{F_y}{A} \qquad \text{(S2)}$$

Where:

$\sigma_y$ : Yield strength, in Pascals (Pa) or megapascals (MPa).

$F_y$: Yield load, the force at which the material begins to yield, in Newtons (N).

$A$ : Cross-sectional area of the sample, in square meters ($m^2$) or square millimeters ($mm^2$).



FigureS2 DMA curves of PP and PP/AOHBP films