Supplementary Information (SI) for Journal of Materials Chemistry A.

Heterodimensional “Nebula-Cluster” Hybrids from MOFs and BP Nanosheets: Towards Multifunctional, Flame Retardant, and Closed-Loop Recyclable Flexible Foam

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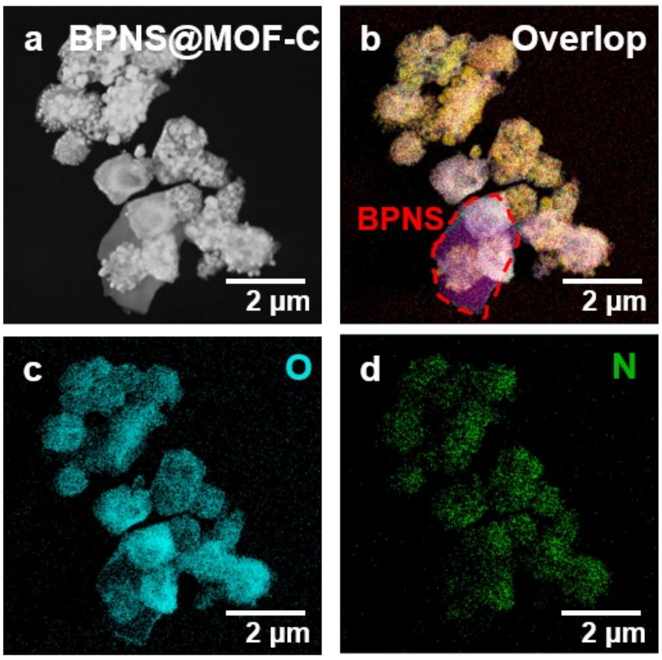

Figure S1. HAADF-STEM image and corresponding elemental (O, N) mapping results of BPNS@MOF-C.

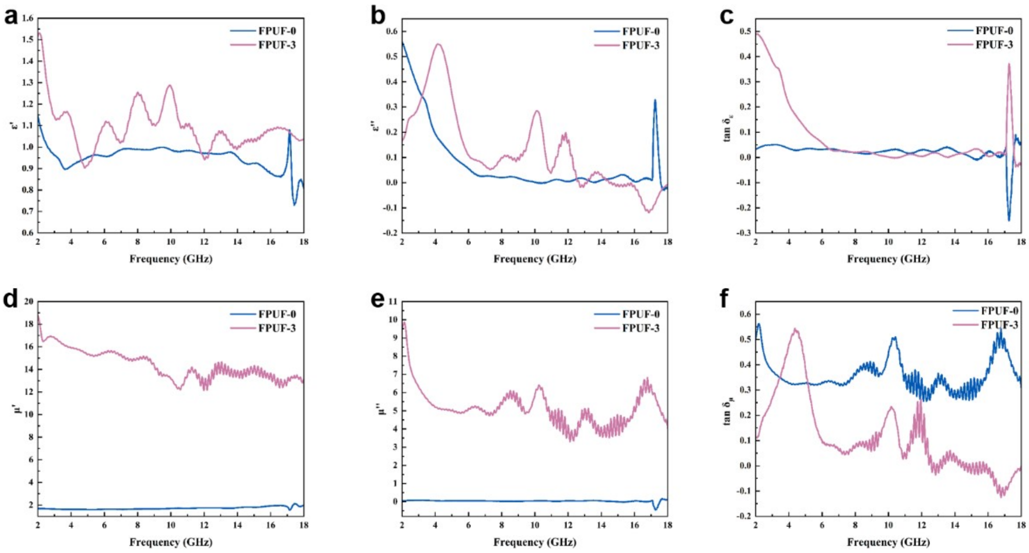

Figure S2. (a) ε′, (b) ε″, (c) μ′, (d) μ″, (e) tan δ_e, (f) tan δ_m curves of FPUF-0 and FPUF-3.

Figure S3. Ambient noise levels in simulated environments without FPUF-3: electric drilling, wall demolition, glass friction and aircraft engine operation.

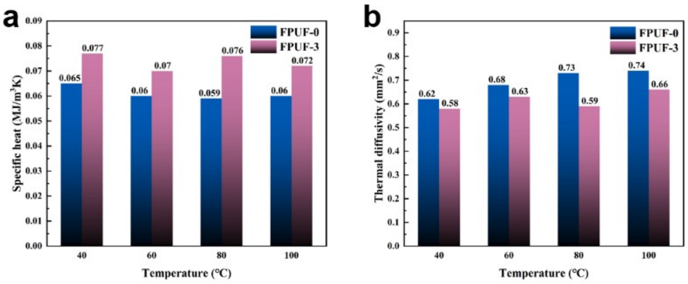

Figure S4. (a) Specific heat and (b) thermal diffusivity of FPUF-0 and FPUF-3.

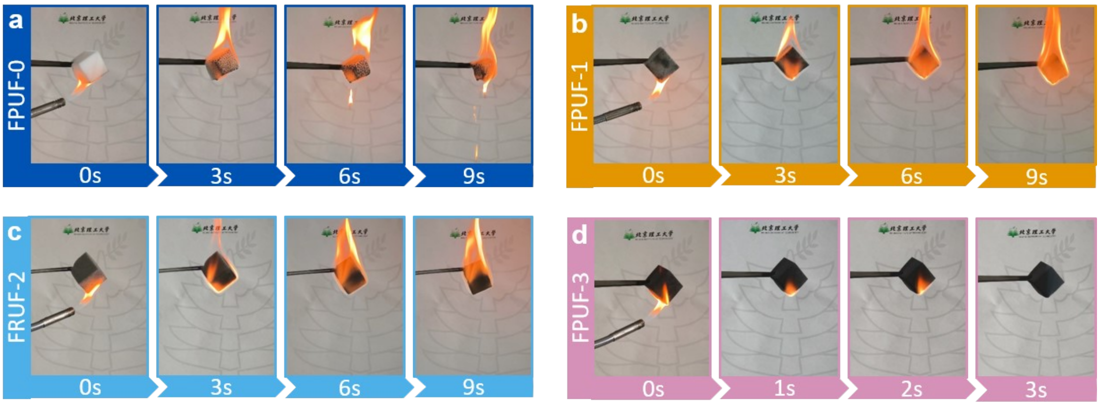

Figure S5. Direct combustion tests (25°C, ambient air) of FPUF.

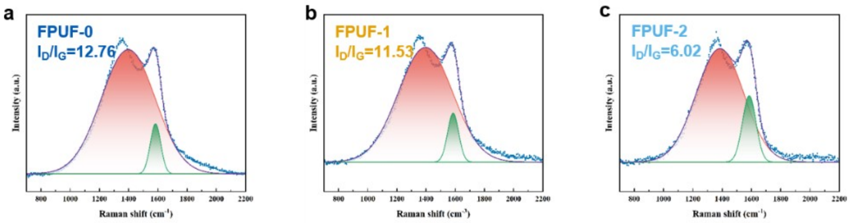

Figure S6. Raman spectra of FPUF-0, FPUF-1 and FPUF-2 after cone calorimetry tests.

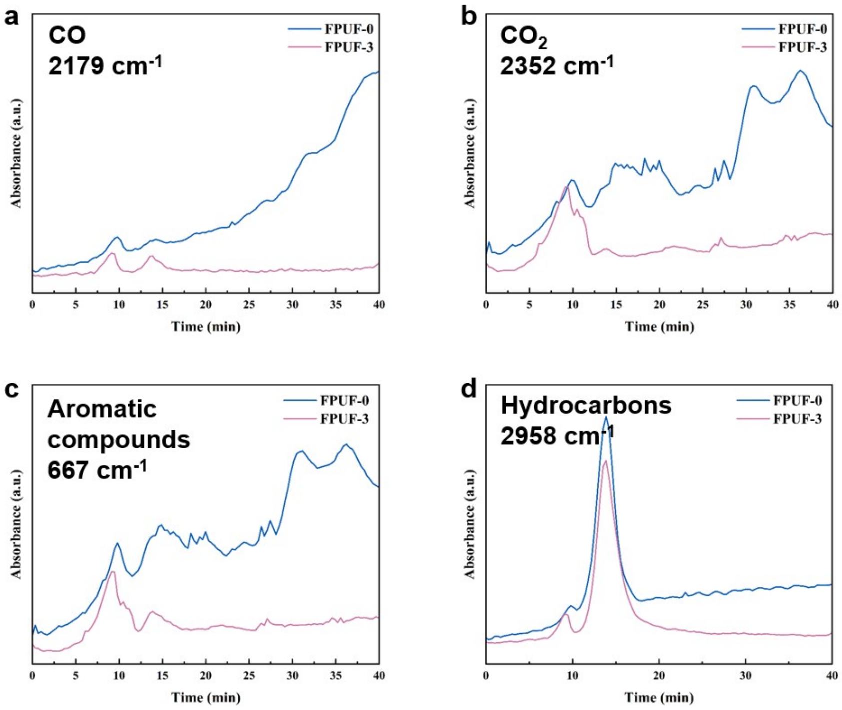

Figure S7. (a) CO, (b) CO_2, (c) aromatic compounds and (d) hydrocarbons of FPUP-0 and FPUF-3.

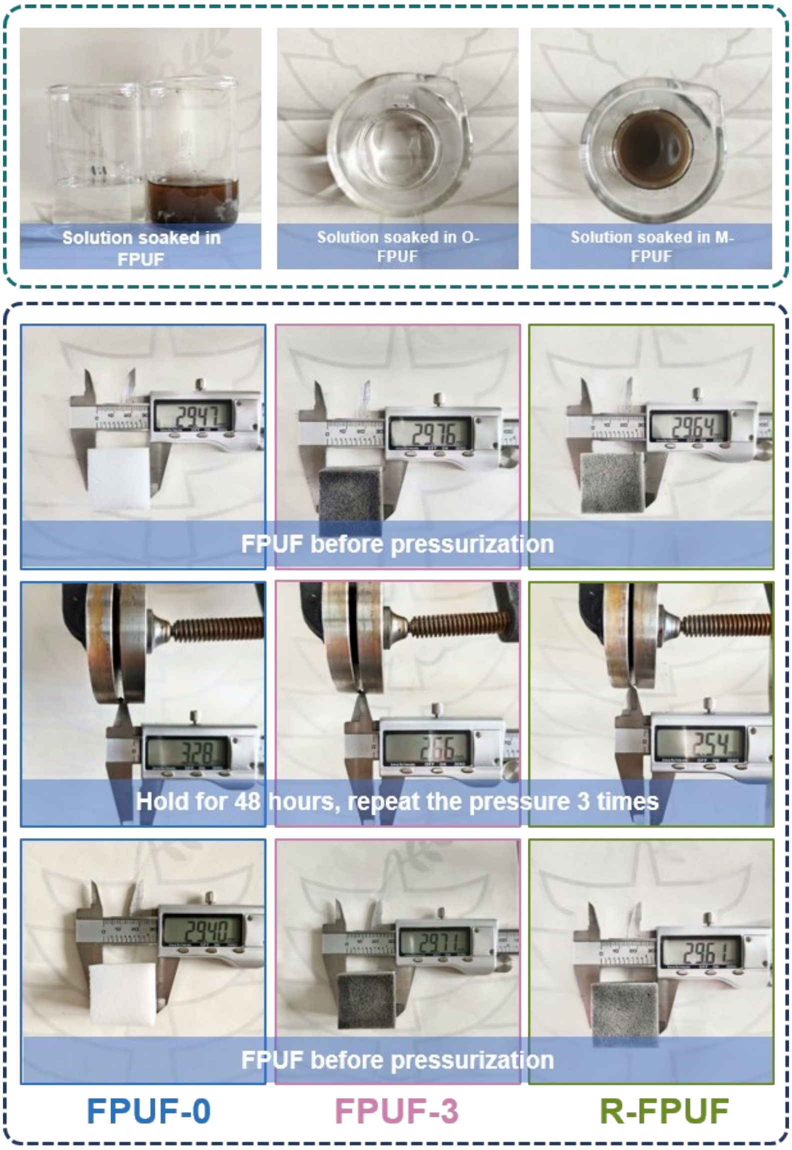

Figure S8. FPUF-3 recovery treatment and compression–recovery process of FPUF-0, FPUF-3 and R-FPUF under 90% strain.

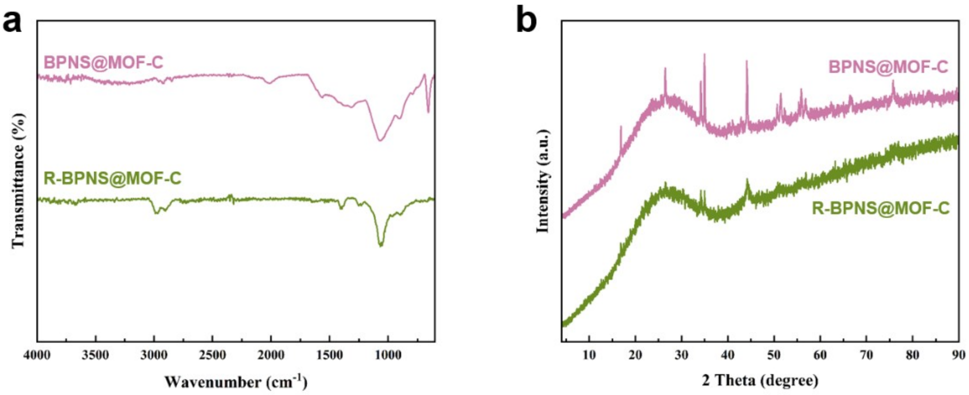

Figure S9. FT-IR spectra and XRD patterns of BPNS@MOF-C and R-BPNS@MOF-C.

4 Experimental

4.1 Materials

Cobalt(II) nitrate hexahydrate ($Co(NO_3)_2·6H_2O$, 99.0%), 2-methylimidazole (2-MeIM, 98.0%), 3,5-diamino-1,2,4-triazole (daTA, 98.0%), Ferrocene (Fc, 98.0%), Ferrocenecarboxaldehyde (Fc-CHO, 99.0%) were all provided by Beijing Tongguang Fine Chemicals Company. Anhydrous methanol (CH_3OH, AR), absolute ethanol (C_2H_5OH, AR), acetic acid (≥99%), N-methylpyrrolidone (NMP, AR) , deionized water (H_2O, AR) and Chitosan (MW=10000) were provided by Macklin Biochemical Co., Ltd. Flexible polyurethane foam (FPUF) was provided by Guangzhou Jintian Spomnge Products Co., Ltd. All chemicals were of analytical grade and were used as received without further purification.

4.2 Preparation of ZIF-67

In a typical synthesis, 2.91 g of $Co(NO_3)_2·6H_2O$ and 3.28 g of 2-MeIM were each dissolved in 100 mL of methanol. The 2-MeIM solution was then added to the cobalt nitrate solution. The combined solution was stirred for 1 hour and allowed to sit undisturbed for 24 hours. The resulting purple precipitate was collected by centrifugation, washed several times with ethanol, and dried at 60 °C for 12 hours.

4.3 Preparation of NH_2-ZIF

NH_2-ZIF was synthesized using the following method: Disperse 0.5 g of ZIF-67 in 50 mL of methanol using ultrasonic dispersion in a water bath. Then, add 50 mL of methanol solution containing 0.5 g of daTA and stir the resulting combination at room temperature for 24 hours. Subsequently, centrifuge to collect the precipitate, wash it multiple times with methanol, and dry at 60 °C for 12 hours.

4.4 Preparation of Fc-ZIF

Fc-ZIF was synthesized using the following method: Disperse 0.5 g of NH2-ZIF in 50 mL of methanol using an ultrasonic water bath. Then, add 50 mL of methanol solution containing 1 g of Fc-CHO and stir for 10 minutes. Subsequently, stir under an oil bath at 60 °C for 12 hours. After cooling to room temperature, centrifuge to collect the precipitate, and wash multiple times with methanol. Dry at 60 °C for 12 hours.

4.5 Preparation of BPNS

0.5 g of BP was dispersed in 500 mL of NMP and sonicated for 6 hours using an ultrasonic cell disruptor (1200 W) under ice bath conditions. The resulting dispersion was first centrifuged at 3000 rpm for 10 minutes to collect the supernatant. Subsequently, the supernatant was further centrifuged at 9000 rpm for 10 minutes to isolate the precipitate. The obtained solid was washed three times with deionized water and ethanol, respectively, and finally dried in a vacuum oven at 60 °C for 12 hours.

4.6 Preparation of BPNS@MOF-C

1 g of Fc-ZIF was weighed into a clean alumina crucible, which was then placed in a tube furnace. Under a nitrogen atmosphere, the temperature was raised to 600 °C at a heating rate of 10 °C/min and held for 3 h. After natural cooling, MOF-C was obtained. Then, 80 mg of MOF-C and 10 mg of BPNS were added to a beaker containing 20 mL of ethanol and sonicated for 30 min. The product was collected by centrifugation and dried in a vacuum oven at 60 °C for 12 h to obtain a BPNS/MOF-C mixture. Subsequently, 1 g of this mixture was placed in a crucible and heated in the tube furnace under nitrogen atmosphere to 300 °C at a rate of 10 °C/min, held for 1 h, and allowed to cool naturally to yield BPNS@MOF-C.

4.7 Preparation of CS- BPNS@MOF-C coated FPUF

The commercially available FPUF was first washed with deionized water and dried for further use. Subsequently, BPNS@MOF-C was dispersed in an aqueous solution of 1% (v/v) acetic acid, followed by ultrasonication for 30 minutes. The cleaned FPUF was immersed into the resulting dispersion and repeatedly compressed to ensure thorough infiltration. After 12 h of immersion, the FPUF was removed and freeze-dried at –50 °C for 48 h.

4.8 Characterization

Fourier transform infrared (FTIR) spectroscopy was obtained on an NLCOLET 6700 IR spectrometer in the range of 400-4000 cm^{-1}. X-ray diffraction (XRD) patterns were collected on a Rigaku MiniFlex 600 Powder X-ray Diffractometer using Cu-Kαradiation at 40 kV and 15 mA (2θ range from 2° to 90° with a step size of 10°/min). The crystal structure of the phases was analyzed using Raman spectroscopy (Thermo

DXR2). X-ray photoelectron spectroscopy (XPS) data were characterized on a Thermo ESCALAB 250Xi. The morphology and microstructure of all samples were observed by a field emission transmission electron microscope (TEM, Tencnai G2 F30 S-TWIN, FEI, USA) and a scanning electron microscope (SEM, SU8020, Hitachi, Japan) equipped with energy dispersive X-ray detector (EDX), respectively. The nitrogen sorption isotherms and pore size distribution were collected on a 3H-2000PS2 analyzer apparatus (BeiShiDe, Beijing) using the Brunauer-Emmett-Teller (BET) method. The data of thermogravimetric analysis (TGA) was obtained on a Netzsch 209 F1 thermal analyzer in the range from 40 to 900°C with a heating rate of 10 °C/min under a nitrogen flow. Thermogravimetric analysis-Fourier transform infrared (TGA-FTIR) was measured by a TGA/DSC 3+ (Mettler-Toledo, Zurich, Switzerland) and an iS50 FTIR spectrometer (Thermo Fisher) in the range from 40 °C to 900 °C with a heating rate of 20 °C/min in a nitrogen atmosphere. Cone calorimeter data were collected on an FTT Cone Calorimeter (Phoenix Instruments Co. Ltd. Suzhou China) according to ISO 5660 under a heat flux of 35 kW/m^2 with the specimen dimensions of 100 × 100 × 5 mm^3. Based on the coaxial transmission/reflection mode, the electromagnetic parameters were measured using the vector network analyzer (Agilent N5222A) in the frequency range of 2-18 GHz. An infrared imaging system (FLIR) was used to record the heat transfer process of the samples on the heating platform. The thermal conductivity of the samples was measure dusing a thermal constants analyzer (Hot Disk TPS2500S, Sweden). The sound absorption performance of the materials was evaluated using a transfer function sound absorption coefficient measurement system (B&K 4206).