Supporting Information

Synergistic effects of flame-retardants on the flammability and foamability of PS foams prepared by supercritical carbon dioxide foaming

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1. Dynamic strain sweep spectra

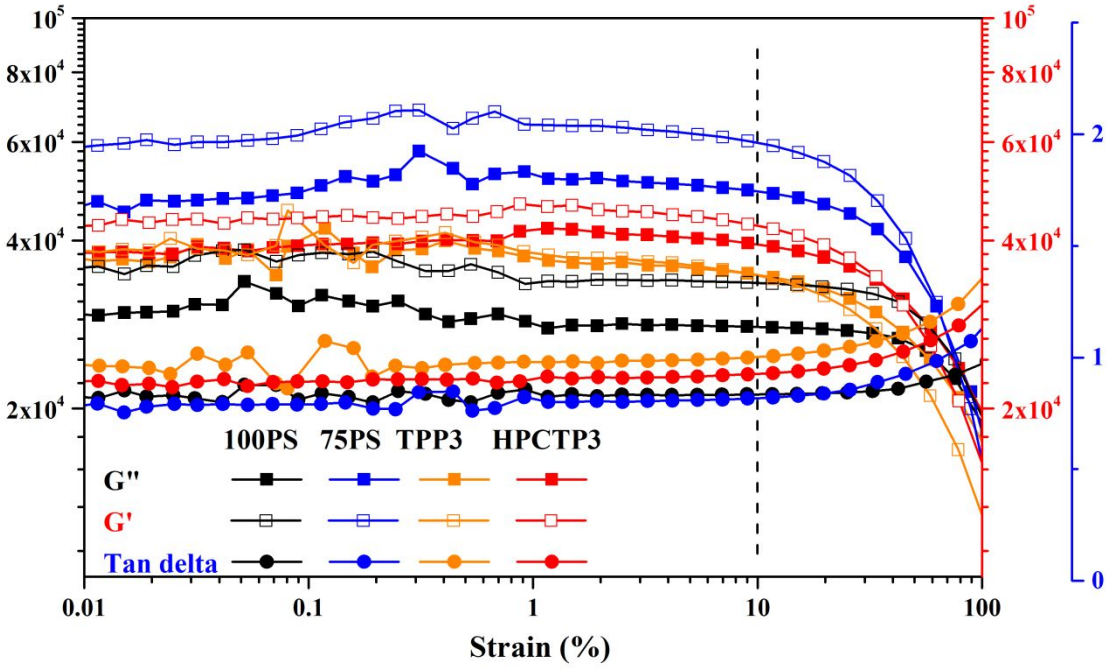

Figure S1 The dynamic strain sweep spectra of 100PS/75PS/TPP3/HPCTP3 composites at 180 °C and 1Hz.

The dynamic strain sweep was adopted to determine the linear viscoelastic region (LVER) of PS composites and the test results were shown as Figure S1. PS and PS composites show the linear viscoelastic region when the strain is lower than 10%. PS and its composites are with high loss modulus (G") and storage modulus (G'), and their order of magnitude is 10^4 (Pa). Neat PS and PS composites in the linear viscoelastic region shows a solid-like behavior (the value of G' is larger than that of G") [1], and the loss tangent (tan δ = G"/G') of 100PS, 75PS, TPP3 and HPCTP3 composites are 0.84, 0.82, 0.98 and 0.91, separately. The loss tangent is a characteristic value to evaluate viscoelastic behavior, and the low values of tan δ ($0.1 < \tan \delta < 1$) of PS composites indicate a predominantly elastic behavior at 180 °C. The addition of TPP and HPCTP can act as the plasticizer and leads to the increase of tan δ, which illustrate that the

viscous behaviors of TPP3 and HPCTP3 composites are enhanced than 75PS composite.

2. MP and EG TGA curves

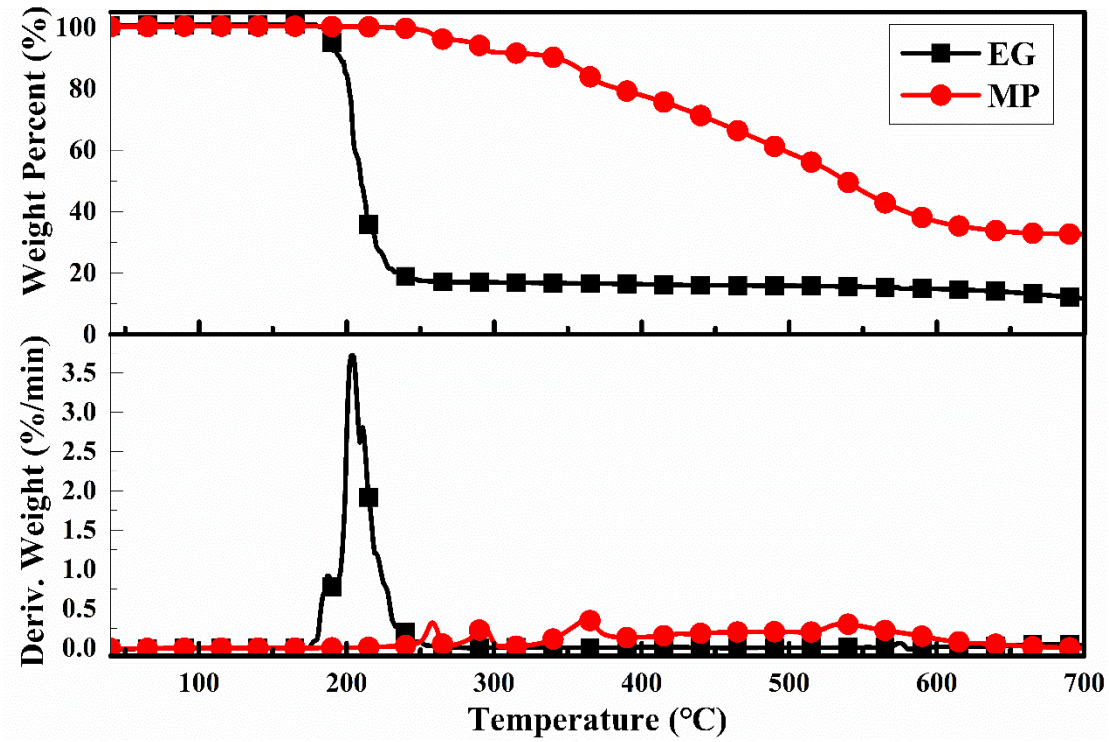

Figure S2 MP and EG* TGA curves under air atmosphere at a heating rate of 10 °C/min.

*The char yield of EG is for reference only, because expanded graphite is with large volume and may fall out of the pan.

As shown in Figure S2, 5% mass loss temperature of MP is 285 °C. The first two decomposition steps (dehydration process) of MP cause 8.43 wt% mass loss, which start from about 200 to 308 °C. As reported in the literature [2], melamine pyrophosphate and melamine polyphosphate are generated during these two steps. In the third decomposition step (308 - 388 °C), melam ultraphosphate, ammonium polyphosphate and other ultraphosphate structure substance were formed by the decomposition of melamine polyphosphate, which are with a mass loss of 12.27 wt% and will release water, ammonia and melamine. The fourth and fifth steps take place from 388 to 700 °C

with 46.79 wt% mass loss, which are attributed to the thermal degradation of ultraphosphate. Finally, the stable residue left at 700 °C is 32.71 wt%. As for EG, its 5% mass loss temperature is 190 °C, this is due to the redox process between H_2SO_4 and the graphite, which releases the blowing gases (CO_2, SO_2 and H_2O). The porous, large volume and worm-like expanded graphite is formed by the expansion and exfoliation along the c-axis of the graphite crystal [3].

Reference:

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2. Laoutid, F.; Bonnaud, L.; Alexandre, M.; Lopez-Cuesta, J.-M.; Dubois, P., New prospects in flame retardant polymer materials: from fundamentals to nanocomposites. *Materials Science and Engineering: R: Reports* **2009,** *63* (3), 100-125.

3. Ye, L.; Meng, X. Y.; Liu, X. M.; Tang, J. H.; Li, Z. M., Flame-retardant and mechanical properties of high-density rigid polyurethane foams filled with decabrominated dipheny ethane and expandable graphite. *J. Appl. Polym. Sci.* **2009,** *111* (5), 2372-2380.