Supporting Information

Green, tough and highly efficient flame-retardant rigid polyurethane foam enabled by double network hydrogel coatings

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Content:

Figures S1–S14 and Tables S1 and S2 (PDF) Movie S1. Scratching test of hydrogel coating by steel needle (MP4) Movie S2. Open fire test of hydrogel-coated RPUF (MP4)

Movie S3. Fire-resistance test of PAAm-PDA hydrogel (MP4)



Figure S1. The FTIR spectra of PAAm, PDA and PAAm-PDA₈ hydrogels.



Figure S2. High-resolution XPS spectra of C 1s region for the (a)PAAm and (b) PAAm-PDA

hydrogels.



Figure S3. The demonstration of the PAAm-PDA hydrogel recovered its initial shape after releasing the compression load.



Figure S4. Optical images of the RPUF and hydrogel-coated RPUF after the compression tests.



Figure S5. Photographs of hydrogel-coated substrate after adhesion test.



Figure S6. Microscope images of scratching test by a steel needle to illustrate mechanical robustness of the hydrogel coating.



Figure S7. Water capture capacity test of PAAm-PDA/CaCl₂ hydrogel in the atmospheric environment (T: 25 °C and RH: 60%).



Figure S8. The SEM images of the surface char of (a) uncoated RPUF, (b) PAAm coated RPUF, (c) PAAm-PDA coated RPUF, and (d) PAAm-PDA (Dry) coated RPUF after the open fire tests.



Figure S9. Photographs of the cross-sectional of PAAm-PDA coated RPUF (a) before and (b) after open fire test.



Figure S10. Photographs of the surface of PAAm-PDA coated RPUF (a) before and (b) after exposure to open fire for (b) 10 s and (c) 20 s.



Blistering

Bursting

Charring

Figure S11. The three combustion stages of the hydrogel-coated substrate during the cone

calorimetry.



Figure S12. Photographs of (a) the experimental set-up and (b) the front view during the test.



Figure S13. Experimental data and fitting curves of heat-transfer tests for hydrogels with different water content and thickness.



Figure S14. The demonstration of the burning process of PAAm-PDA hydrogel by putting it on top of a hotplate at 300 °C.

| Samples | AAm | BIS | APS | TMEDA | DA/AAm | Water |
|-----------------------|------------|------|------|-------|---------|---------|
| | (g) | (mg) | (mg) | (μL) | (wt. ‰) | (wt. %) |
| PAAm | 2.5 | 5 | 50 | 20 | 0 | 80 |
| PAAm-PDA ₂ | 2.5 | 5 | 200 | 20 | 2 | 80 |
| PAAm-PDA ₄ | 2.5 | 5 | 200 | 20 | 4 | 80 |
| PAAm-PDA ₈ | 2.5 | 5 | 200 | 20 | 8 | 80 |

Table S1. The compositions of various hydrogel coatings

 Table S2. Comparison of the cone results and mechanical strength of the hydrogel-coated RPUF

 with results of fire-retardant RPUF reported elsewhere [1-9]

| Samples | TTI (s) | ∆ Mean | Δ TSP | △ Compression | Ref. |
|----------------------|---------|--------|--------|----------------------|-----------|
| | | HRR | | strength | |
| DPPM (25)-RPUF | | -44.4% | | -25% | [1] |
| TSPB (30)-RPUF | | | | -31.6% | [2] |
| RPUF/HDPCP25 | 3 | | -10.4% | -9.4% | [3] |
| 2%PRPUF/15%EG | 6 | | | +4.4% | [4] |
| RPUF-5 | 2 | | | -28.8% | [5] |
| 20A/20Z-RPUF | 3 | | +6.3% | +7.1% | [6] |
| RPUF-PMAPP25 | 7 | | | -23.6% | [7] |
| RPUF-100 | 3 | | +33.3% | -36% | [8] |
| RPUF/PBM-m1.0 | 5 | -13.2% | +2.1% | +20% | [9] |
| Hydrogel-coated RPUF | 36 | -39.7% | -42.2% | +31.8% | This work |

Note: -- stands for no data.

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