Supporting Information

Enhancing Performance in Blue Quantum-Dot Light-emitting Diodes by Using a Deep HOMO Carbazol-Phenyl-Triazine Small Molecule Blended Hole Transporting Layer

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Experimental Section

Materials: Clevios PVP AL 4083 poly(3,4-ethylenedioxythiophene)/poly(styrenesulfonate) (PEDOT:PSS) was purchased from Heraeus and Xi'an Polymer Light Technology Co. Poly(9-vinylcarbazole) (PVK), MoO_3, and chlorobenzene were purchased from Sigma-Aldrich. Blue CdSe/CdS/ZnS QDs was supplied by Guangdong Poly Optoelectronics Co., Ltd. N-octane was obtained from Aladdin Reagent (Shanghai) Co., Ltd. All reagents were used directly without further purification after purchase.

Synthesise of ZnO nanoparticles: The mixture of Zinc acetate (1 g, 4.56 mmol) and Ethanol absolute (37.5 mL) was placed in a three-necked round-bottomed flask and heated to 85 °C under nitrogen atmosphere using an oil bath until the zinc acetate was dissolved. Subsequently, KOH (0.5 g, 8.9 mmol) was mixed in Ethanol absolute (5 mL) and dissolved by ultrasonic shaking. Then the dissolved Ethanol absolute solution of KOH was slowly added dropwise to Ethanol absolute solution of Zinc acetate, and then kept heated for 30 min until the solution in the three-necked flask appeared milky white. After the reaction was completed it was cooled to room temperature and poured into a 150 mL flask, about 2 times the volume of n-hexane was added while stirring, and allowed to stand for 12 h in a refrigerator. After the white product was completely settled, the supernatant was poured off and the process of settling and standing was repeated again. Then the settled supernatant was poured off and the remaining suspension was evenly distributed in a centrifuge tube and centrifuged at 6000 r/min for 4 min. The supernatant in the centrifuge tube was poured off and the remaining solids in the tube were blown dry with a nitrogen gun to remove as much of the residual n-hexane as possible. Finally, the precipitate was dispersed with high purity ethanol (>99.8%) with the addition of appropriate amount of ethanolamine, which was carried out in an ice water bath. After complete

dispersion, it was centrifuged again and the resulting supernatant was extremely ZnO NPs.[50]

Synthesis of 2,4,6-tris(3-(carbazol-9-yl)- phenyl)-triazine (TCPZ): A mixture of 2,4,6-tris(3-bromophenyl)-1,3,5-triazine (TBrPZ) (0.979 g, 1.8 mmol), carbazole (1.05 g, 6.3 mmol), $PdCl_2$ (28.7 mg, 0.162 mmol), tris(tertbutyl) phosphine (131 mg, 0.648 mmol), and sodium tertbutoxide (0.778 g, 8.1 mmol) in anhydrous o-xylene (100 mL) was stirred at 120 °C for 17 h under nitrogen atmosphere. After cooling to room temperature, the mixture was poured into water and then extracted with chloroform. The combined organic phase was washed with brine and dried over $MgSO_4$. The subjection of the crude mixture to silica gel chromatography (chloroform/n-hexane = 2/1) afforded TCPZ (1.14 g, 79%) as white powders. 1H NMR (500 MHz, CDCl3): d (ppm) 8.93 (t, J = 2.0 Hz, 3H), 8.74 (d, J = 8.0 Hz, 3H), 8.15 (d, J = 8.0 Hz, 6H), 7.78 (d, J = 8.0 Hz, 3H), 7.73 (t, J = 8.0 Hz, 3H), 7.42 (d, J = 8.0 Hz, 6H), 7.33 (t, J = 7.0 Hz, 6H), 7.28 (t, J = 7.0 Hz, 6H). 13C NMR (500 MHz, CDCl3): d (ppm) 171.26, 140.76, 138.26, 137.84, 131.30, 130.30, 128.06, 127.60, 126.08, 123.42, 120.32, 120.11, 109.60. MS (EI): m/z 806 [M+H+] (calcd m/z 804.94). Anal. Calc. for C57H36N6 (%): C, 85.05; H, 4.51; N, 10.44. Found: C, 85.10; H, 4.70; N, 10.14. The results is consistant with published paper. [22]

Device Fabrication: Before the preparation of the corresponding devices, the ITO substrate was respectively cleaned with the electronic cleaning agent, deionized water, acetone, and isopropylalcohol by ultrasonic cleaning for 30 min, followed by drying in the oven at 80 °C, and then treated with oxygen plasma for about 10 min, which was to complete a series of preparation work for the substrate before manufacturing the corresponding devices. First, PEDOT:PSS was uniformly spin-coated onto each ITO substrate at 3000 rpm for 40 s and then annealed on a hot table at 150 °C for 20 min to remove any residual water. All samples were then immediately transferred to a nitrogen glovebox for subsequent spin-coating of multilayer films and deposition of electrodes. To prepare the hole transport layer, PVK and TCPZ were dissolved in chlorobenzene at a concentration of 4 mg mL^{-1}, respectively, filtered and blended at different volume ratios, and then the PVK and TCPZ blends were coated on PEDOT:PSS at a rotational speed of 2000 rpm for 30 s. Finally, the films with PVK and TCPZ deposited by spin coating were annealed on a hot table at 125 °C for 20 min. Then, the QDs EML and ZnO ETL were deposited. N-octane solution of blue QDs (concentration of 10 mg mL^{-1}) was first deposited on the upper layer of TCPZ:PVK film sequentially at 2000 rpm for 30 s, and then the QDs layer was annealed at 105 °C for 20 min, after which the ethanol solution of ZnO nanoparticles (Synthesized in our laboratory at a concentration of 30 mg mL^{-1}) was spin-coated and deposited in the same way as above, except that the annealing temperature of the layer was 105 °C. At last, the samples were moved to a thermal evaporation system, and 110 nm Al electrodes were deposited on the ZnO surface by evaporation with a shadow mask at a vacuum of 4×10^{-4} Pa. The overlap area between the cathode and anode is about 10 mm^2.

Characterizations: The photoelectric characteristics of the QLEDs devices were measured in a nitrogen-filled glove box. The current density and luminance of the device with voltage were tested by the Keithley 2400 source meter and Konica Minolta LS-150 luminance meter, respectively. A Marine optical fiber spectrometer (Maya 2000 Pro) was used to collect the corresponding devices' electroluminescence (EL) spectra. The photoluminescence (PL) spectra of TCPZ: PVK/QDs, PVK/ blue QDs, TCPZ/QDs，and blue QDs films were taken by a Horiba FluoroMax-4 spectrofluorometer. A functional wavelength generator (Rigol DG4162) was used to provide a pulse voltage for the device's transient EL fluorescence lifetime testing. The roughness of the film surface was measured by an atomic force microscope (AFM, LT UHV) from Scienta Omicron in Germany. Shimadzu's X-ray photoelectron spectroscopy (AXIS SUPRA), the UPS test to analyze the energy bands of the semiconductor films and UV-visible absorption spectra were recorded by an HP8453A spectrophotometer. Finally, the Contact Angle Measuring Instrument (SDC-100) is adopted by Dongguan Shengding Precision Instrument Co., the infrared thermal imaging camera is based on the German Instruments testo thermography.
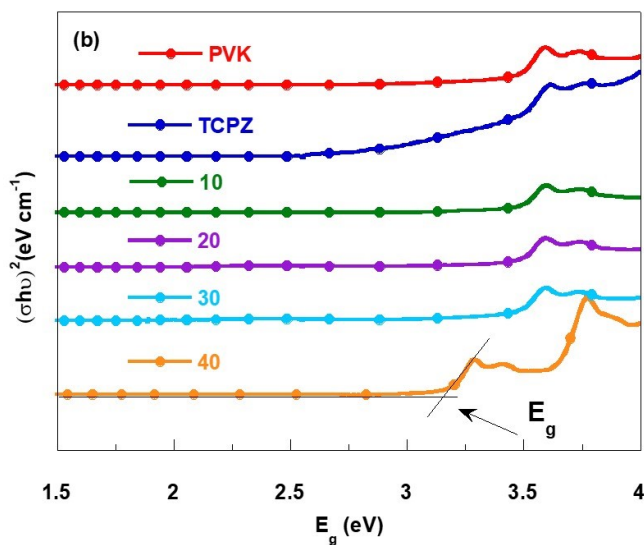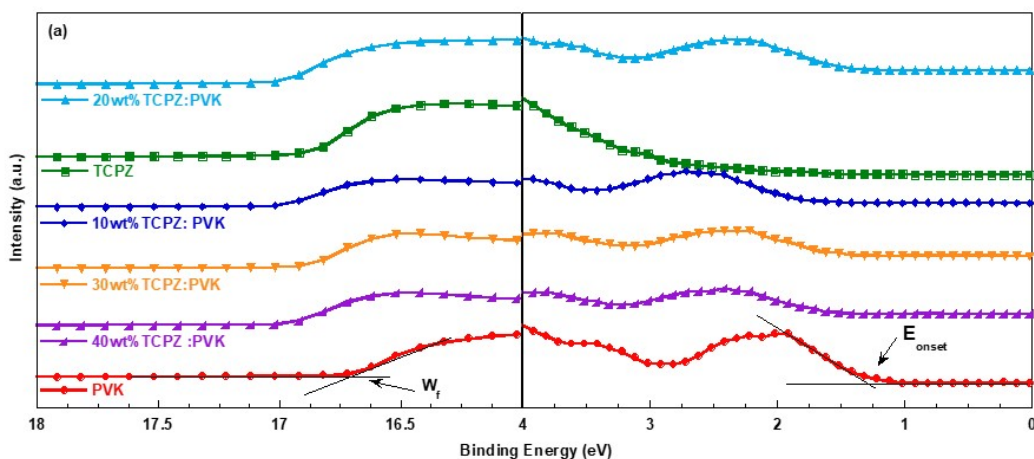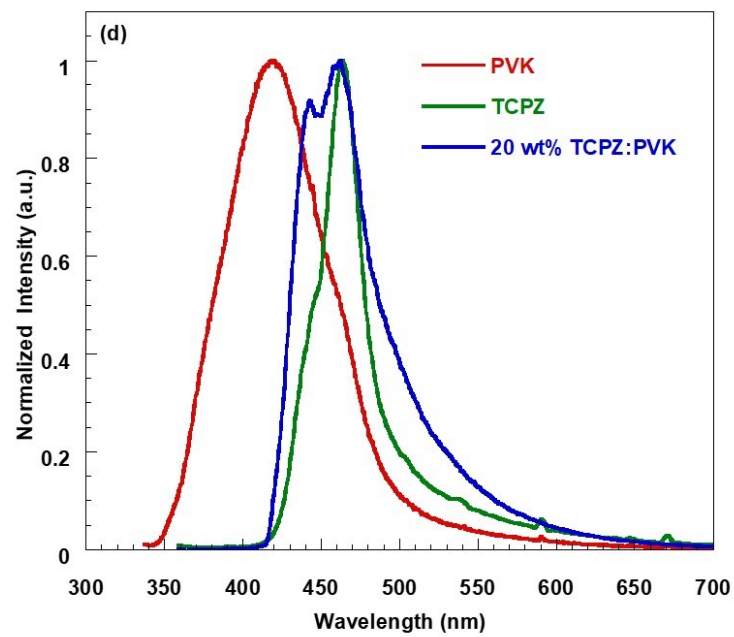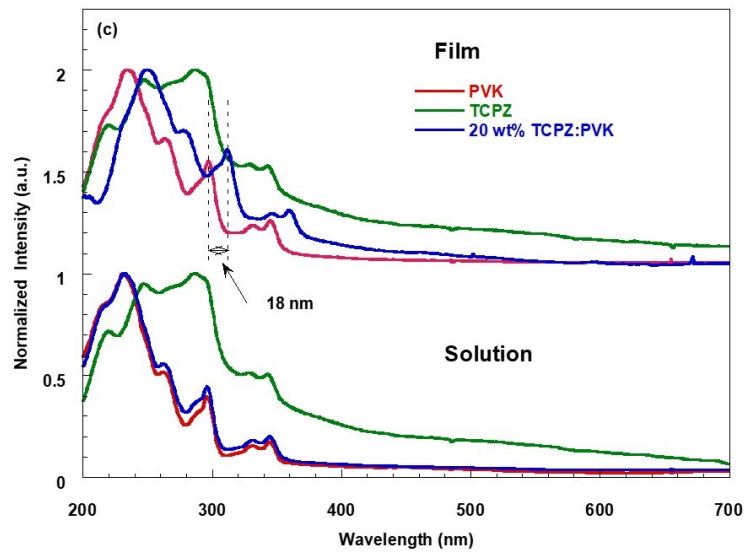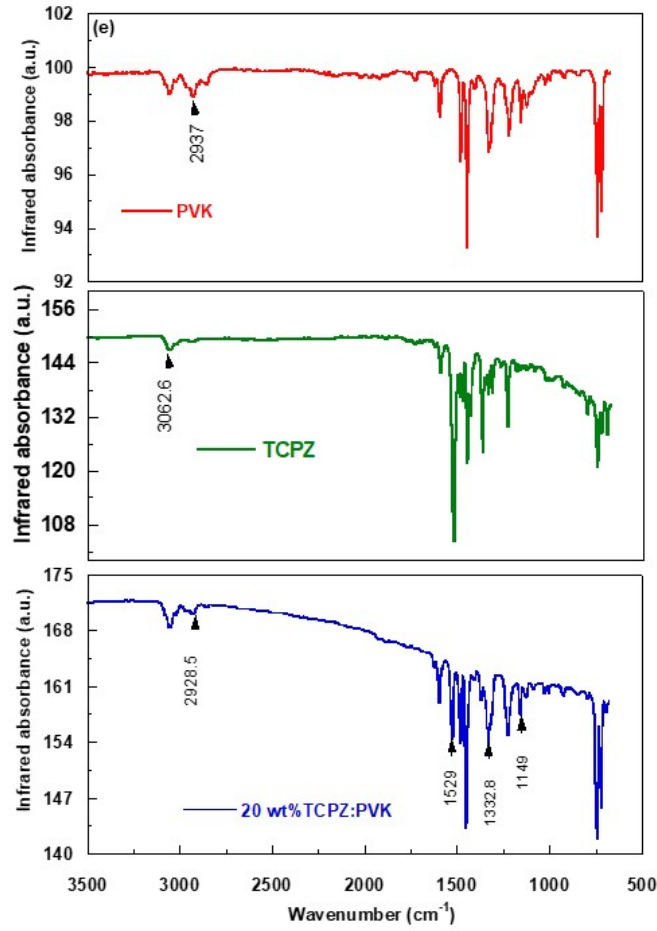
Figure S1 (a) UPS spectrum of PVK, TCPZ, and x wt%TCPZ: PVK films. (b) Band-gap widths were fitted according to absorbed data. (c) UV- visible absorption spectra of PVK, TCPZ, and x wt% TCPZ:PVK films or solutions. (d) PL spectra of PVK, TCPZ, and x wt% TCPZ:PVK films. (e) Infrared spectra of PVK, TCPZ, and 20wt% TCPZ:PVK.

The hole mobility of TCPZ: PVK film layers can be obtained by fitting the space charge-limited current (SCLC) region with Child's-law (the fitted curves are shown in Figure S2), where V_{TFE} is the initial voltage of the trap filling limit.[28-29] From Table S1, the V_{TFE} of TCPZ: PVK is lower than that of neat PVK devices, especially the V_{TFE} of 20 wt% TCPZ: PVK is reduced from 0.72 V to 0.08 V for neat PVK, and the lower V_{TFE} indicates that the concentration of trap density within the film layer is decreased, which is beneficial to enhance the efficiency of the devices, and further revealed that the addition of TCPZ can rise the trap vacancies of PVK and indirectly advance the hole mobility. Next, we use the mobility fitting $J = \dfrac{9\mu_e \varepsilon_0 \varepsilon_r V^2}{8d^3}$ to calculate the hole mobility for different blending ratios, and $\varepsilon_0, \varepsilon_r, \mu_e, d$ denote the vacuum permittivity, relative permittivity, carrier mobility, and film thickness, respectively.
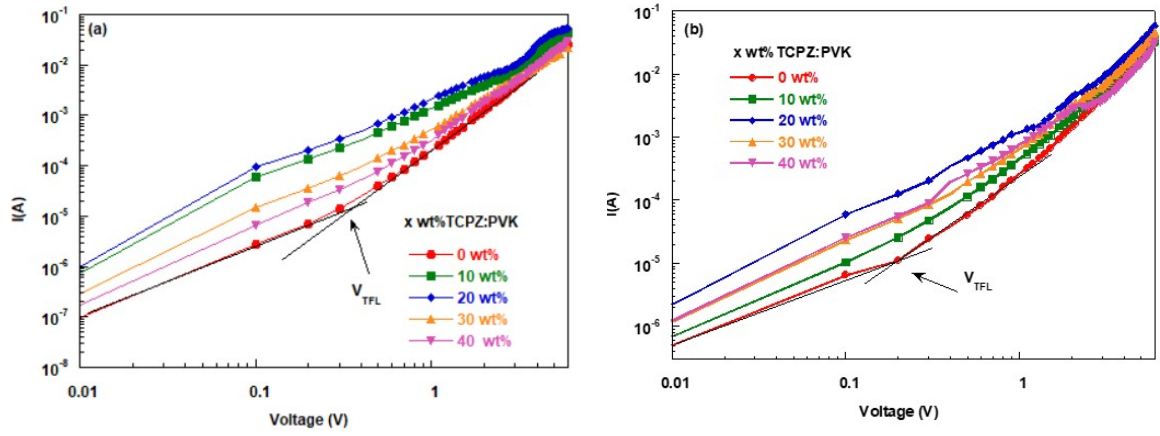
Figure S2 Child's -law fitting space charge limited current (SCLC) region for single-hole devices structured as (a) Glass/ITO/PEDOT:PSS (40 nm)/x wt% TCPZ:PVK (40 nm)/blue CdSe/Cds/ZnS QDs (30 nm) /MoO$_3$ (10 nm)/Ag (100 nm), and (b) Glass/ITO/PEDOT:PSS (40 nm)/x wt% TCPZ:PVK (40 nm)/ MoO$_3$ (10 nm)/Ag (100 nm).

Table S1 Filling limits the initial voltage and mobility of the hole trap with different blending ratios (the structure is Glass/ITO/PEDOT:PSS (40 nm)/x wt% TCPZ:PVK (40 nm)/blue CdSe/Cds/ZnS QDs (30 nm) /MoO$_3$ (10 nm)/Ag (100 nm)).

x wt%TCPZ:PVK	V$_{TFL}$	µ$_e$
x=0	0.72 V	2.32×10^{-7}
x=10	0.11 V	8.39×10^{-6}
x=20	0.08 V	1.07×10^{-5}
x=30	0.29 V	4.77×10^{-6}
x=40	0.41 V	8.06×10^{-7}

Table S2 Filling limits the initial voltage and mobility of the hole trap with different blending ratios (the structure is Glass/ITO/PEDOT:PSS (40 nm)/x wt% TCPZ:PVK (40 nm) /MoO$_3$ (10 nm)/Ag (100 nm)).

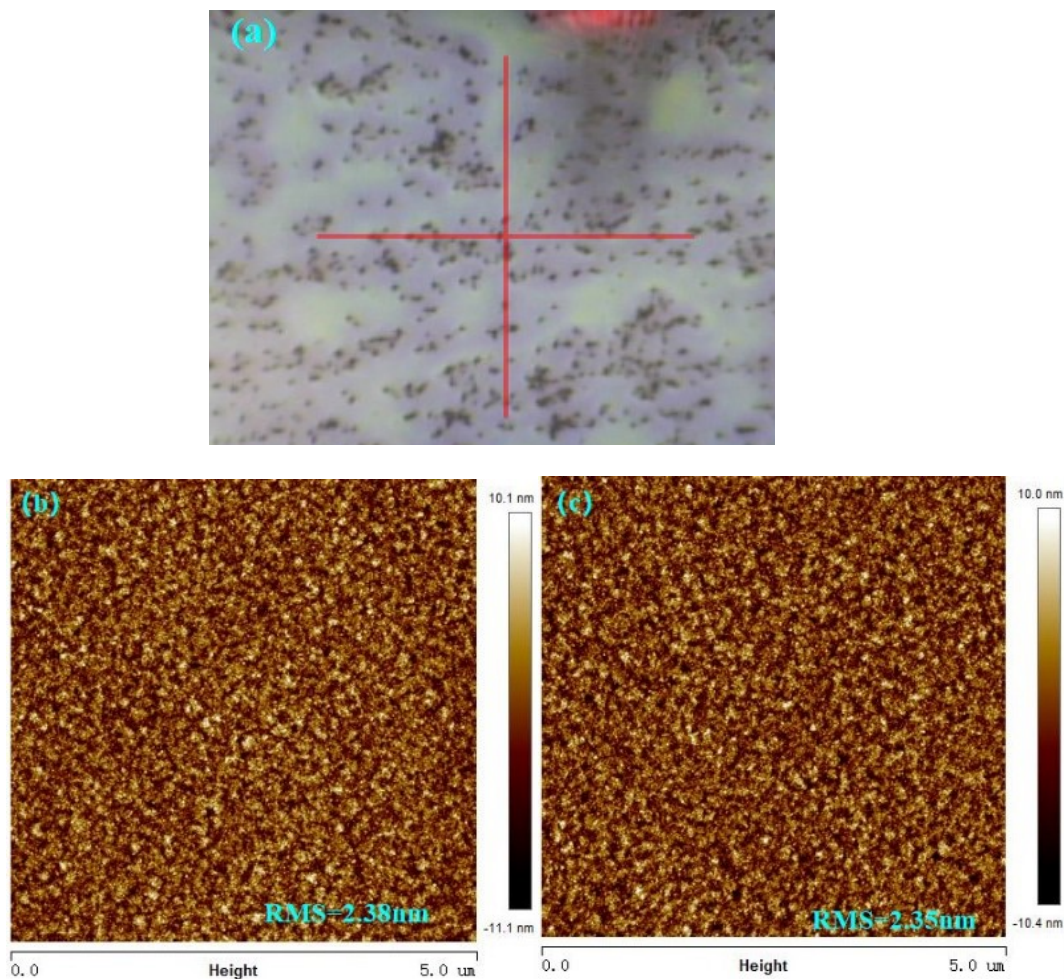| x wt%TCPZ:PVK	V$_{TFL}$	µ$_e$
x=0	0.41 V	1.31×10^{-6}
x=10	0.20 V	1.02×10^{-5}
x=20	0.09 V	9.34×10^{-5}
x=30	0.30 V	6.68×10^{-6}
x=40	0.38 V	3.17×10^{-6}

Figure S3 (a) 30 wt%TCPZ:PVK film, (b) PVK/blue QDs double-layered film, and (c) 20 wt% TCPZ:PVK /blue QDs double-layered film.
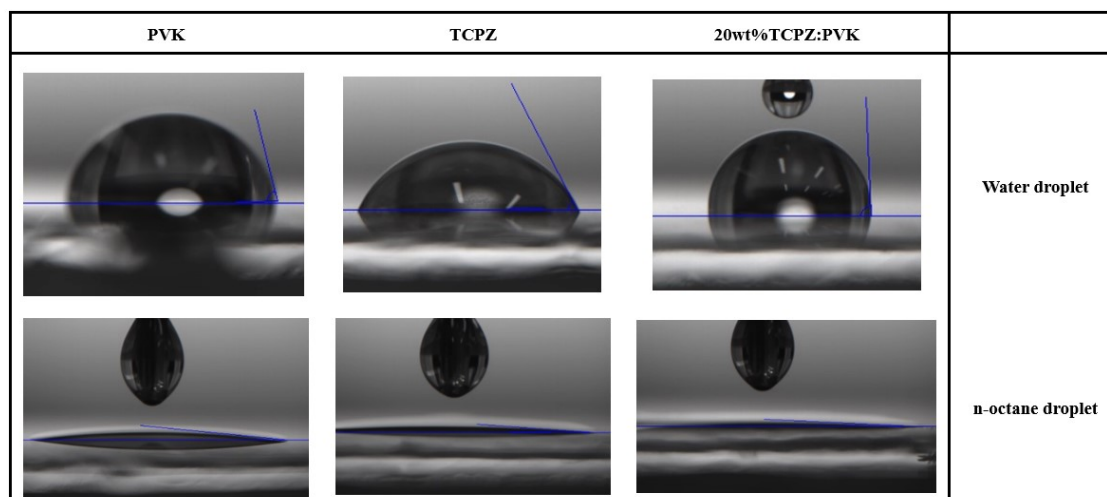

Figure S4 Water and n-octane solvent testing of surface contact angle of PVK, TCPZ, and 20 wt%TCPZ:PVK films.

Table S3 Values of contact angle and surface energy for different films.

Film	water contact angle	n-octane contact angle	surface energy (mN/m)
PVK	78.046°	5.618°	43.49
TCPZ	61.446°	3.74°	43.55

| 20wt%TCPZ:PVK | 87.553° | 2.759° | 45.17 |