

Supporting Information

Efficient Electrocatalytic Upgradation of Furan-Based Biomass: Key Roles of a Two-Dimensional Mesoporous Poly(*m*-phenylenediamine)-Graphene Heterostructure and a Ternary Electrolyte

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1. Materials and methods

Materials. 1-Butyl-3-methylimidazolium tetrafluoroborate (BmimBF₄, 98%), 1-butyl-3-methylimidazolium nitrate (BmimNO₃, 98%), 1-butyl-3-methylimidazolium hexafluorophosphate (BmimPF₆, 98%), 1-butyl-3-methylimidazolium acetate (BmimCH₃COO, 98%), and 1-butyl-3-methylimidazolium dihydrogen phosphate (BmimH₂PO₄, 98%) were purchased from the Centre of Green Chemistry and Catalysis, Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences. Phytic acid (PA, 50 wt%), m-phenylenediamine (99%), and ammonium persulfate (99%) were supplied by J&K Scientific Co., Ltd. Poly(ethylene oxide)₂₀-*block*-poly(phenylene oxide)₇₀-*block*-poly(ethylene oxide)₂₀ (P123, 99%), furfuryl alcohol (98%), furfural (98%), and 5-methylfurfural (98%) were obtained from Sinophar Chemical Reagent Co., Ltd. Nafion D-521 dispersion (5% w/w in water and 1-propanol, ≥0.92 meg/g exchange capacity), Nafion N-117 membrane (0.180 mm thick, ≥0.90 meg/g exchange capacity), and Toray Carbon Paper (CP, TGP-H-60, 19×19 cm) were purchased from Alfa Aesar China Co., Ltd. All of the reactants were purified before use.

Preparation of 2D mesoporous electrocatalysts. Taking the meso-PA/PmPD/GO as an example, First, GO was synthesized according to the modified Hummers method.¹ P123 (0.4 g) was dissolved in deionized water (40 mL). Then, 8 mL GO aqueous solution (0.82 mg mL⁻¹) was added into the mixture solution. After mild stirring at 35°C for 1 h, m-phenylenediamine (50 mg) and PA (33 μL) were added into the solution. The mixture was stirred continuously for 1 h, then 20 mL aqueous solution of ammonium persulfate (10 mg mL⁻¹) was added dropwise to initiate the cross-linking polymerization of *m*-phenylenediamine and PA. Afterwards, the resultant black precipitate was washed with ethanol and water to remove the P123 template. The product is denoted as meso-PA/PmPD/GO.

For the synthesis of meso-PmPD/GO, the procedures were similar to the meso-PA/PmPD/GO except the addition of PA. For non-PA/PmPD/GO, the difference is without the addition of P123 as the template in the reaction system.

Preparation of the electrodes. The obtained electrocatalyst (10.0 mg) was suspended in 1 mL ethanol with 20 μL Nafion D-521 dispersion (5 wt%) to form a homogeneous ink with the assistance of ultrasound. Nafion D-521 is employed for the dispersion of electrocatalysts and their adhesion of electrocatalysts on CP; it does not affect the electrocatalytic process.² After that, the ink

was spread onto carbon paper (CP) (1 cm×1 cm). The CP was pretreated by the calcination at 500 °C in the air before use. Finally, the electrode was dried under room temperature.

2. Characterizations.

Transmission electron microscope (TEM, JEOL-2100) and field emission transmission electron microscope (FETEM, TALOS F200X) with an accelerating voltage of 200 kV were used for the TEM characterizations. TEM samples were prepared by dropping a drop of the sample dispersion containing ethanol on copper grids, followed by air drying at room temperature. Scanning electron microscopy (SEM) experiment was conducted on a Hitachi S-4800 scanning electron microscope operated at 15 kV. SEM samples were prepared by dropping a drop of the sample dispersion containing ethanol on silicon wafers, followed by air drying at room temperature. Powder X-ray diffraction (XRD) patterns were collected on a Rigaku D/max-2500 X-ray diffractometer using Cu K α radiation ($\lambda = 0.154$ nm). Ultraviolet photoelectron spectroscopy (UPS) was conducted on the Scienta R3000 spectrometer system using HeI 21.22 eV as exciting source with energy resolution of 50 meV. ^1H NMR spectra were recorded on a Bruker Avance III HD 400 MHz NMR spectrometer. Fourier transform infrared spectra (FTIR) were conducted using a Nicolet 6700. UV-Vis spectra were carried out on a UV/EV300 spectrophotometer (Thermo Fisher). Atomic Force Microscopy (AFM) images were collected on Nanonavi E-Sweep (Japan). Raman spectroscopy was conducted on a Senterra R200-L (Bruker Optics). Gas chromatography-mass spectrometer (GC-MS) measurement (Agilent 5975C-7890A) was used for the qualitative analysis of products. Variable temperature FTIR spectra were recorded using a Nicolet 6700 at a heating rate of 2 °C/min. Prior to the testing, the mixture of furfuryl alcohol and the 2D catalyst were mixed with KBr by grinding and pressing into pellets.

Linear sweep voltammetry (LSV) measurements. An electrochemical workstation (CHI 660E, Shanghai CH Instruments Co., China) was employed in all the electrochemical experiments in this work. Linear sweep voltammetry (LSV) measurements were conducted on an H-type cell, which was separated by Nafion 117 membrane. A three-electrode system including working electrode, a platinum gauze auxiliary electrode, and a Ag/Ag $^+$ (0.01 M AgNO $_3$ in 0.1 M TBAP-MeCN) reference electrode was adopted. Prior to experiment, the air in the electrolyte was removed by bubbling with N $_2$ for 30 minutes. All the potentials reported were converted from vs. Ag/Ag $^+$ to vs. NHE by adding a value of 0.8 V.³

Cyclic voltammetry (CV) measurements. CV measurements were similar to LSV. An H-type cell and three-electrode system were employed. Prior to experiment, the air in the electrolyte was removed by bubbling with N₂ for 30 minutes.

Electrochemical impedance spectroscopy (EIS). EIS was conducted in a single compartment cell with three electrodes, including a working electrode, a platinum gauze auxiliary electrode, and a Ag/Ag⁺ (0.01 M AgNO₃ in 0.1 M TBAP-MeCN) reference electrode. The EIS spectra were collected using the potentiostatic mode at an open circuit potential of 100 kHz to 100 mHz with an amplitude of 5 mV.

Electrochemical oxidations of furfuryl alcohol and furfural. Electrochemical oxidations of furfuryl alcohol and furfural were performed in a typical H-type cell at room temperature (Figure S32). The anodic and cathodic electrolytes were BmimBF₄-MeCN-H₂O and aqueous H₂SO₄ solution (0.5 M), respectively. The amount of electrolyte in each chamber was 5.9 g in all experiments. Prior to electrolysis, the anolyte was bubbled with N₂ gas for 30 min under stirring. Then, furfuryl alcohol (0.45 mmol) or furfural (0.5 mmol) was added into the anolyte. The electrochemical reaction was started at a desired applied potential for 2 h. The liquid product was analyzed by ¹H NMR (Bruker Avance III 400 HD spectrometer) in DMSO-*d*₆ or D₂O. The conversion and selectivity of the reaction were calculated in terms of ¹H NMR spectra.

3. Density functional theory (DFT) calculations.

DFT calculations were carried out using the Gaussian 16 A.03 program.⁴ Geometric optimization, transition state searches, vibrational analysis and a series of single point energy were performed at the B3LYP⁵⁻⁷/6-311G(d,p) level. In this work, structures of transition states were determined by relaxed potential energy surface (PES) scans and transition state searches, then confirmed using frequency calculations and IRC calculations. Solvation Model Based on Density (SMD)⁸ were used in all calculations involved in this work to provide the effect of implicit solvent (dimethyl sulfoxide). Unless otherwise specified, all discussed energies were Gibbs free energies. In the calculation of surface adsorption, considering the periodicity of PmPD in the 2D catalysts, the simplified surface fragments were employed. The PES between furfuryl alcohol and BmimH on the surface of PmPD with/without PA doping was studied. The optimized structures were obtained by potential energy minima. The corresponding coordinates of DFT-computed stationary points were shown in Page S33-S55.

4. Supplementary figures and tables

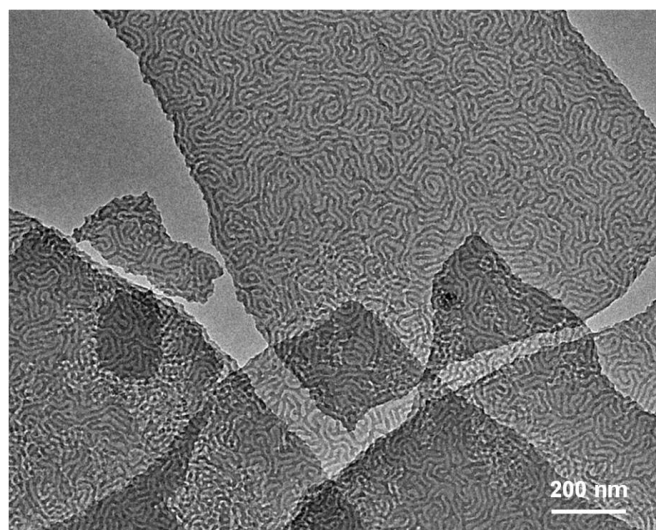


Figure S1. TEM image of the meso-PA/PmPD/GO nanosheets.

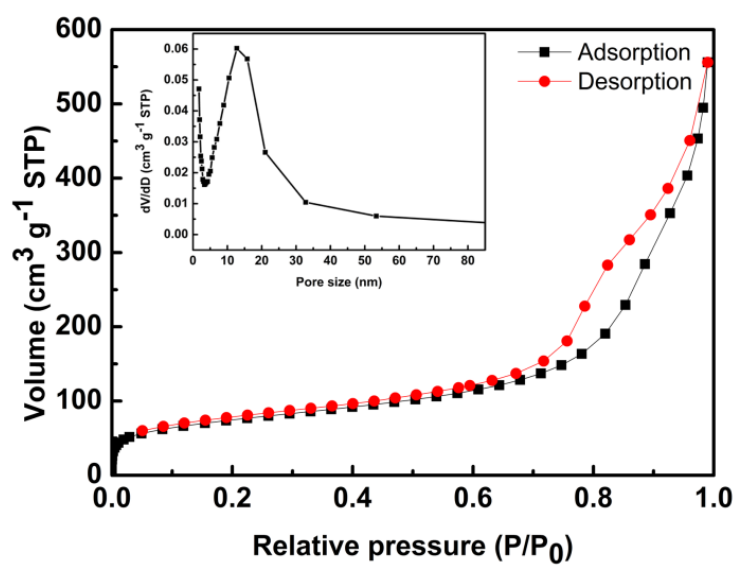


Figure S2. N₂ adsorption-desorption isotherm of the meso-PA/PmPD/GO nanosheets.

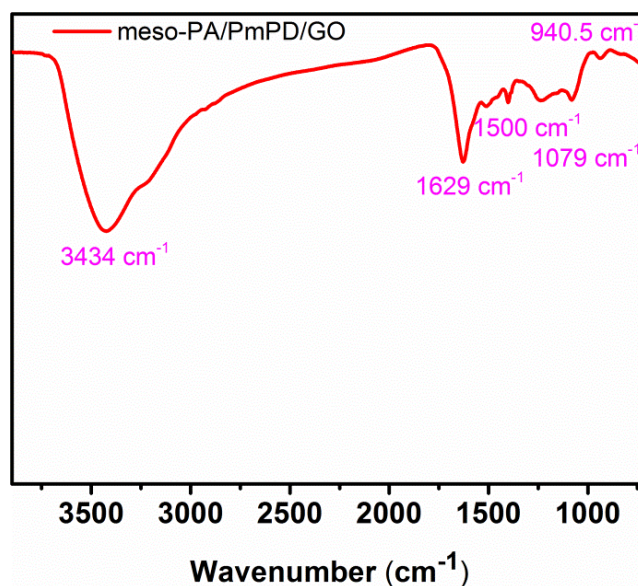


Figure S3. A FTIR spectrum of the meso-PA/PmPD/GO nanosheets.

In the FTIR spectrum, the broad peak at 3434cm^{-1} is ascribed to the stretching vibration of -NH- and -OH. The sharp peaks located at 1629 cm^{-1} and 1500 cm^{-1} are assigned to the stretching vibrations of quinoid imine and benzenoid amine, respectively.⁹ The peaks at 940.5 cm^{-1} and 1079 cm^{-1} are attributed to the stretching vibrations of the phosphate group.^{10, 11} Meanwhile, the FTIR spectrum does not show typical signals attributed to P123, indicating the removal of P123.¹²

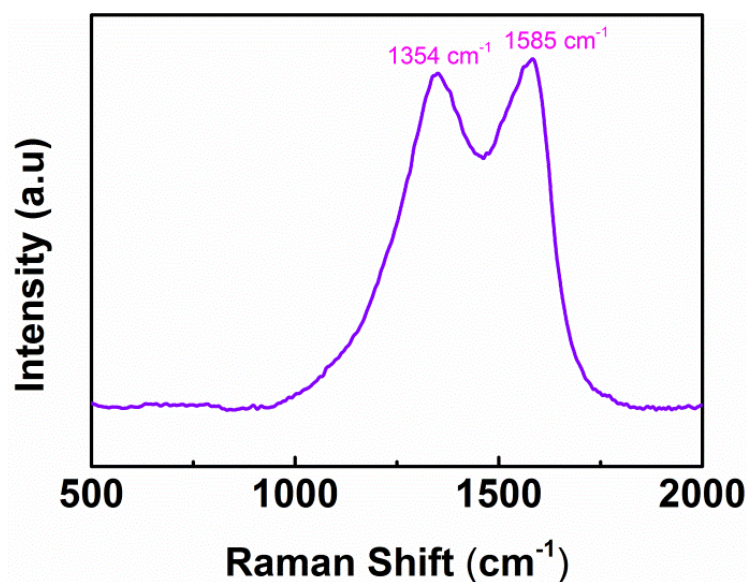


Figure S4. A Raman spectrum of the meso-PA/PmPD/GO nanosheets.

In the Raman spectrum, the two peaks at 1354 cm^{-1} and 1585 cm^{-1} are assigned to the D and G

bands of GO, respectively. The D band corresponds to the carbon atoms with a stretching vibration of sp^3 hybridization, which reflects defects and disorders. The G band corresponds to the stretching vibration of sp^2 -carbon atoms.

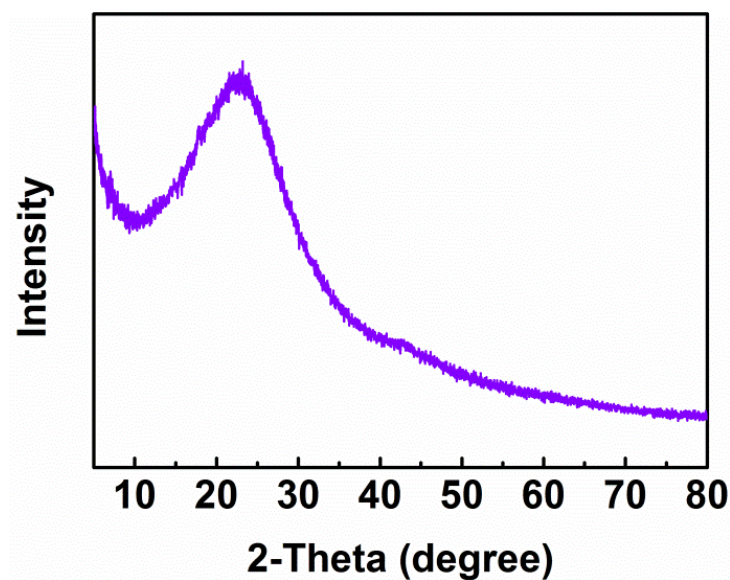


Figure S5. The XRD pattern of the meso-PA/PmPD/GO nanosheets.

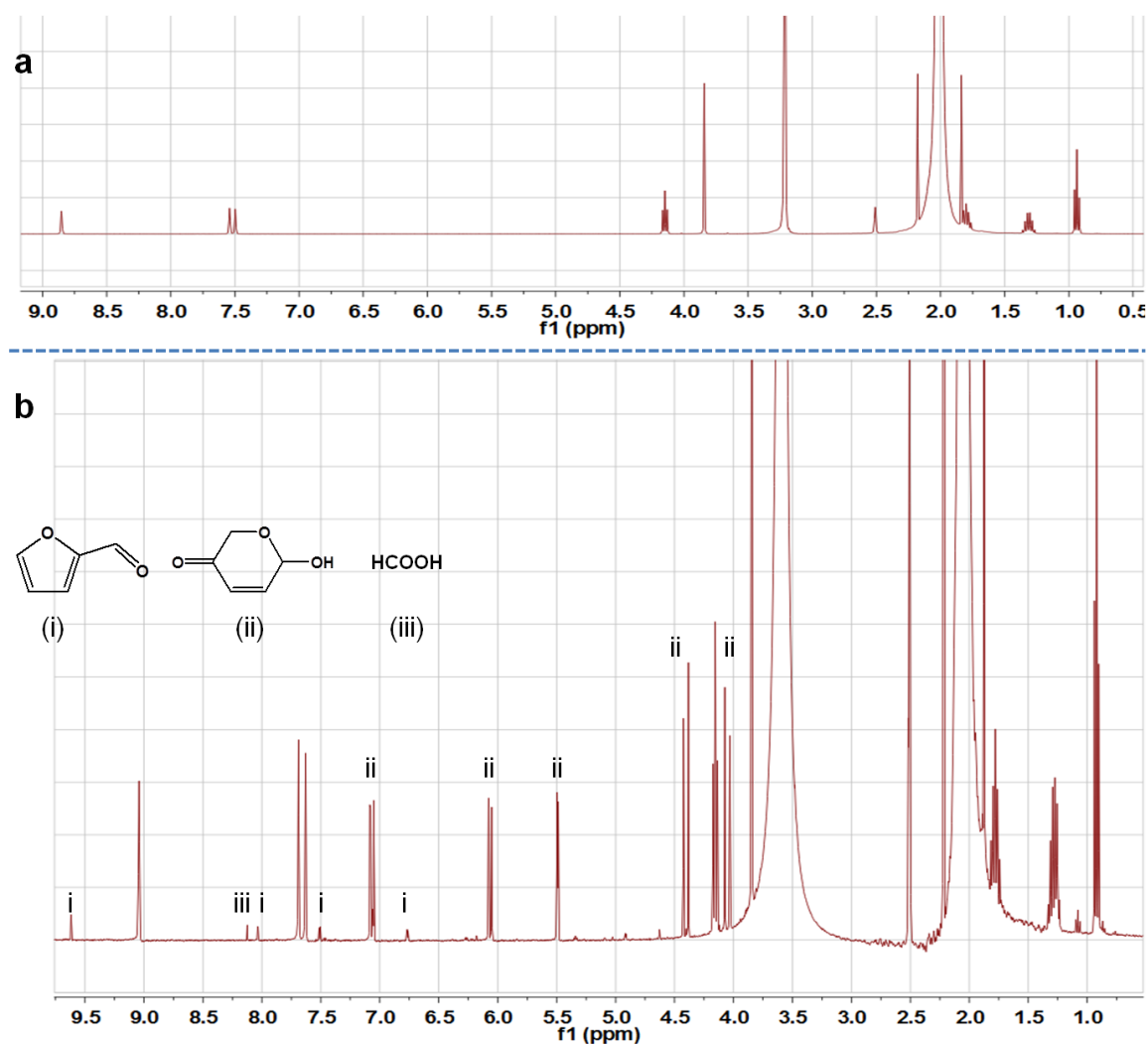


Figure S6. ¹H NMR characterization. (a) ¹H NMR spectra of the ternary electrolyte. (b) ¹H NMR spectra of after the electrochemical oxidation of furfuryl alcohol over the meso-PA/PmPD/GO electrocatalyst in the ternary electrolyte using DMSO-*d*₆ as solvent.

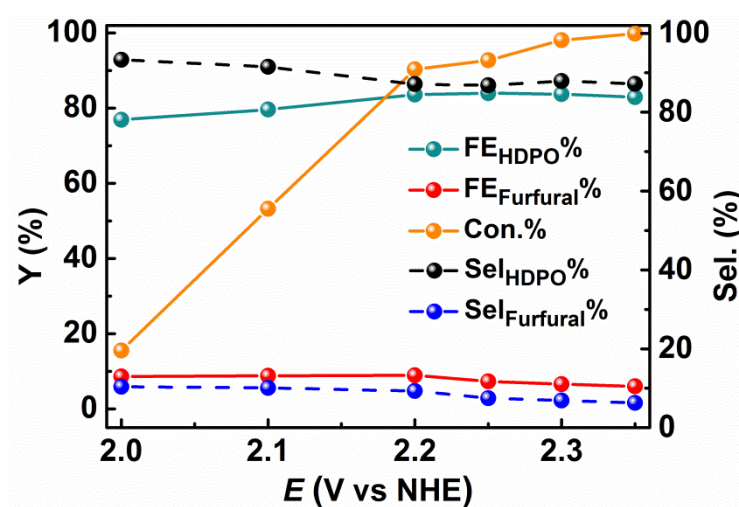


Figure S7. Effect of the potential on the electrochemical oxidation of furfuryl alcohol.

The effect of the potential on the electrochemical oxidation of furfuryl alcohol is presented in Figure S7. At a lower potential (2.0 V vs NHE), a low conversion (15.5%) of furfuryl alcohol is obtained because of the low power. With an increase in the potential, the conversion of furfuryl alcohol increases, whereas the selectivity of HDPO decreases due to the conversion of partial furfuryl alcohol to furfural. In the range of 2.0-2.2 V, the oxidation of furfuryl alcohol to HDPO is dominant. As the potential increases to the range of 2.2-2.35 V, the FE of HDPO starts to reduce, whereas both the selectivity and FE of furfural also decrease. The reason is that the resultant furfural can be partly oxidized to produce HCOOH through the cleavage of the aldehyde group of furfural. Meanwhile, the competitive oxygen evolution reaction *via* the electrolysis of H₂O is accompanied at higher potentials. Considering the balance of the conversion, FE and selectivity, 2.3 V is selected as the optimal potential for the subsequent investigations.

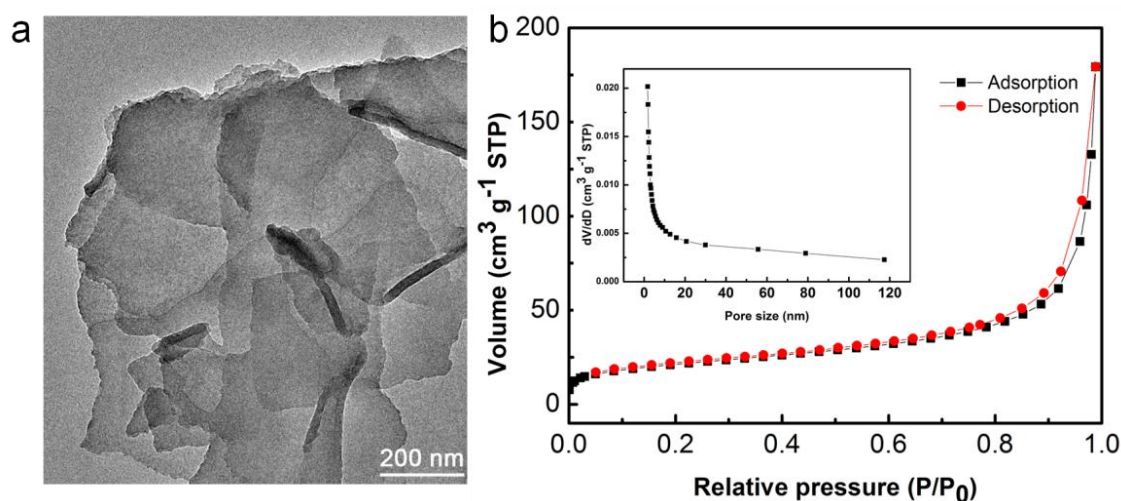


Figure S8. The structural characterizations of non-PA/PmPD/GO. (a) TEM image. (b) N₂ adsorption-desorption isotherm.

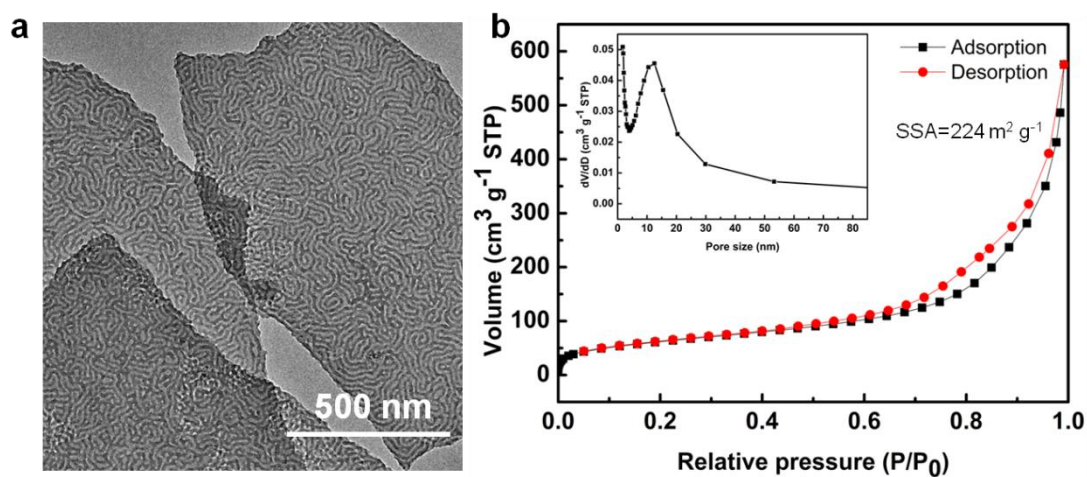


Figure S9. The structural characterizations of meso-PmPD/GO. (a) TEM image. (b) N_2 adsorption-desorption isotherm.

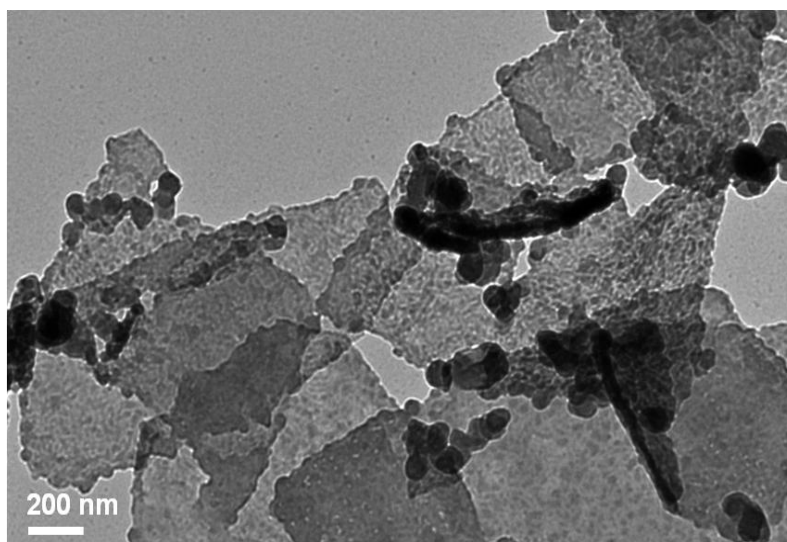


Figure S10. TEM image of meso-PA/PmPD/GO with 19.1wt% PA.

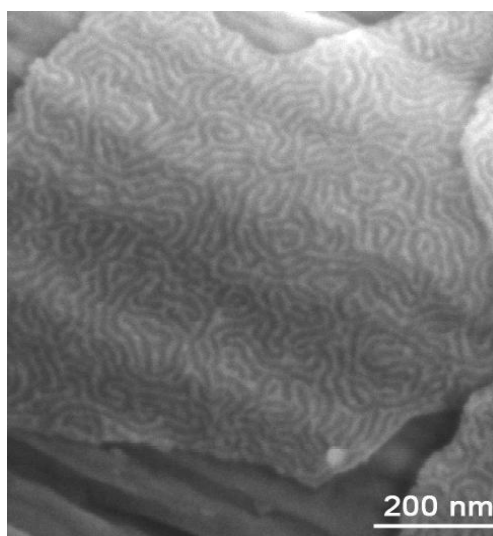


Figure S11. SEM image of the used meso-PA/PmPD/GO nanosheets after the electrochemical oxidation of furfuryl alcohol.

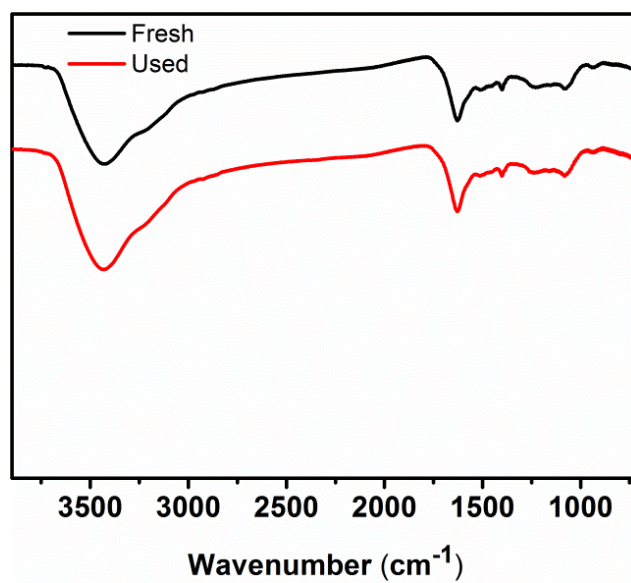


Figure S12. FTIR spectra of the fresh and used meso-PA/PmPD/GO.

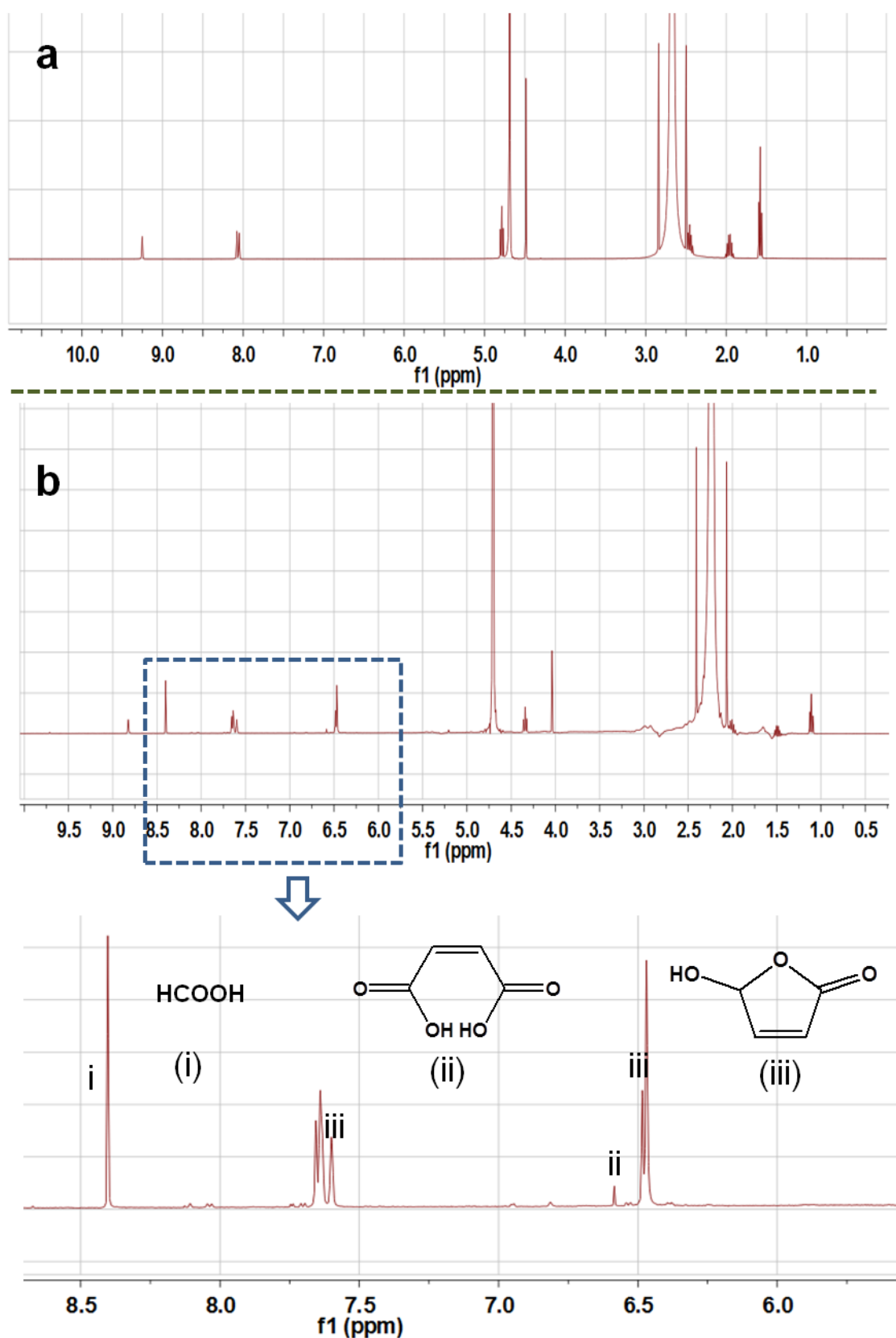


Figure S13. ^1H NMR characterization. (a) ^1H NMR spectra of the ternary electrolyte. (b) ^1H NMR spectra of after the electrochemical oxidation of furfural over the meso-PA/PmPD/GO electrocatalyst in the ternary electrolyte using D_2O as solvent.

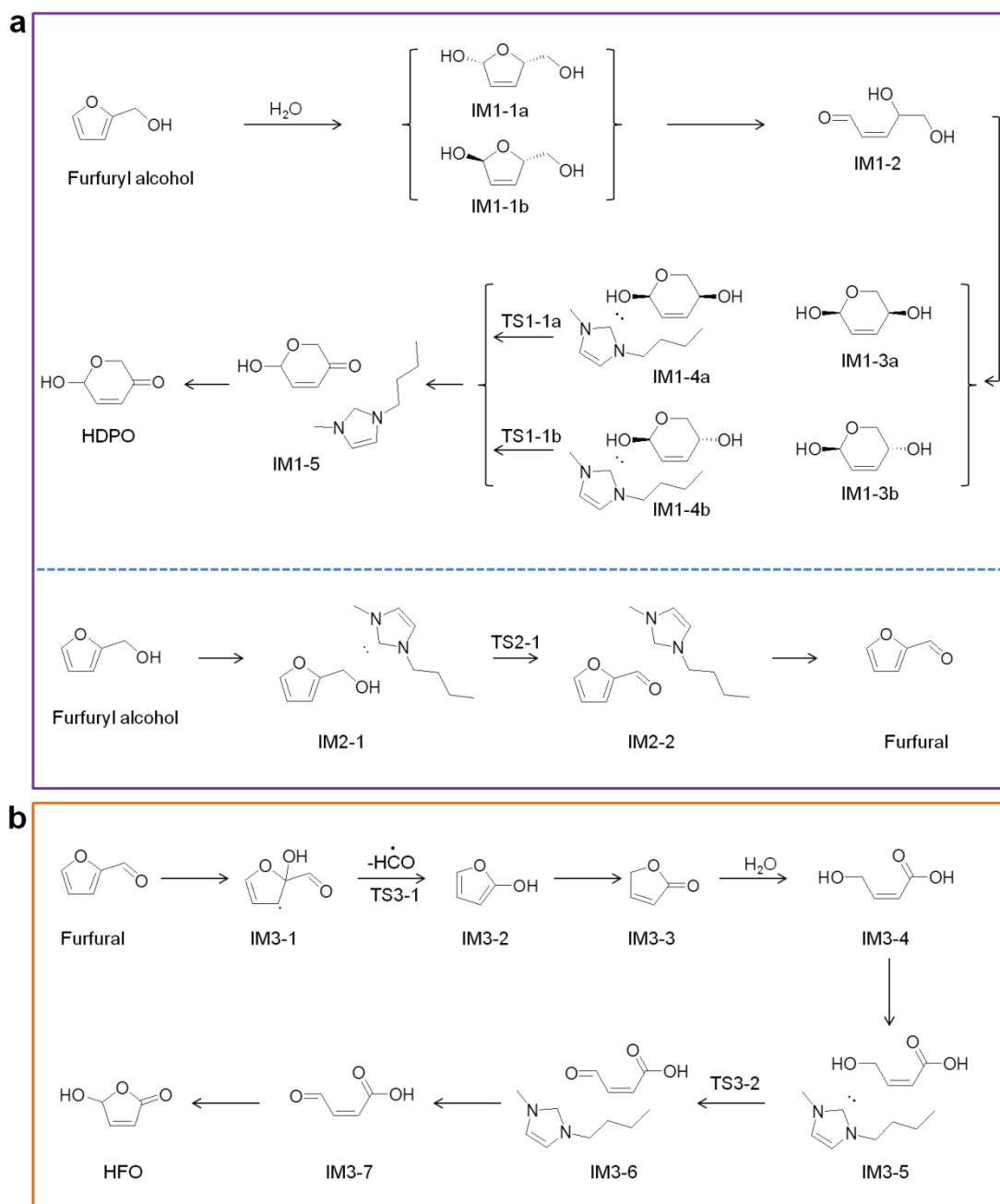
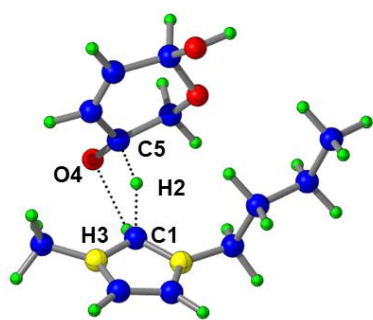
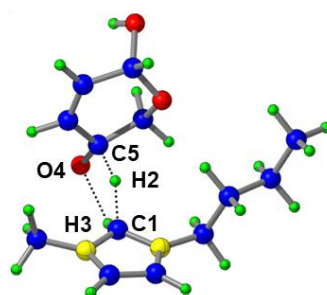


Figure S14. Reaction mechanism of the electrochemical oxidation of furan-based biomass. (a) furfuryl alcohol. (b) furfural.



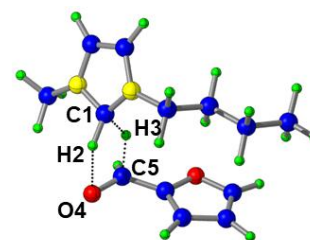
C1-H2: 1.42
C1-H3: 1.10
O4-H3: 2.09
O4-C5: 1.28
C5-H2: 1.42

TS1-1a



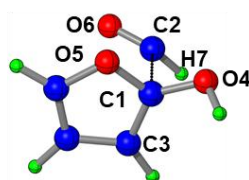
C1-H2: 1.42
C1-H3: 1.10
O4-H3: 2.01
O4-C5: 1.27
C5-H2: 1.43

TS1-1b



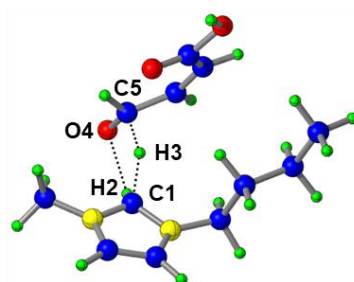
C1-H2: 1.10
C1-H3: 1.42
O4-H2: 1.98
C5-H3: 1.44
O4-C5: 1.27

TS2-1



C1-C2: 2.15
C1-C3: 1.39
C1-O4: 1.35
C1-O5: 1.37
O6-C2-H7: 125.64
C1-O5-C3-O4: 10.81

TS3-1



C1-H2: 1.10
C1-H3: 1.43
O4-H2: 1.98
C5-H3: 1.44
O4-C5: 1.28

TS3-2

Figure S15. 3D structures of the transition states (TS) in the process of electrochemical oxidation furfuryl alcohol and furfural. The bond distances are given in Å.

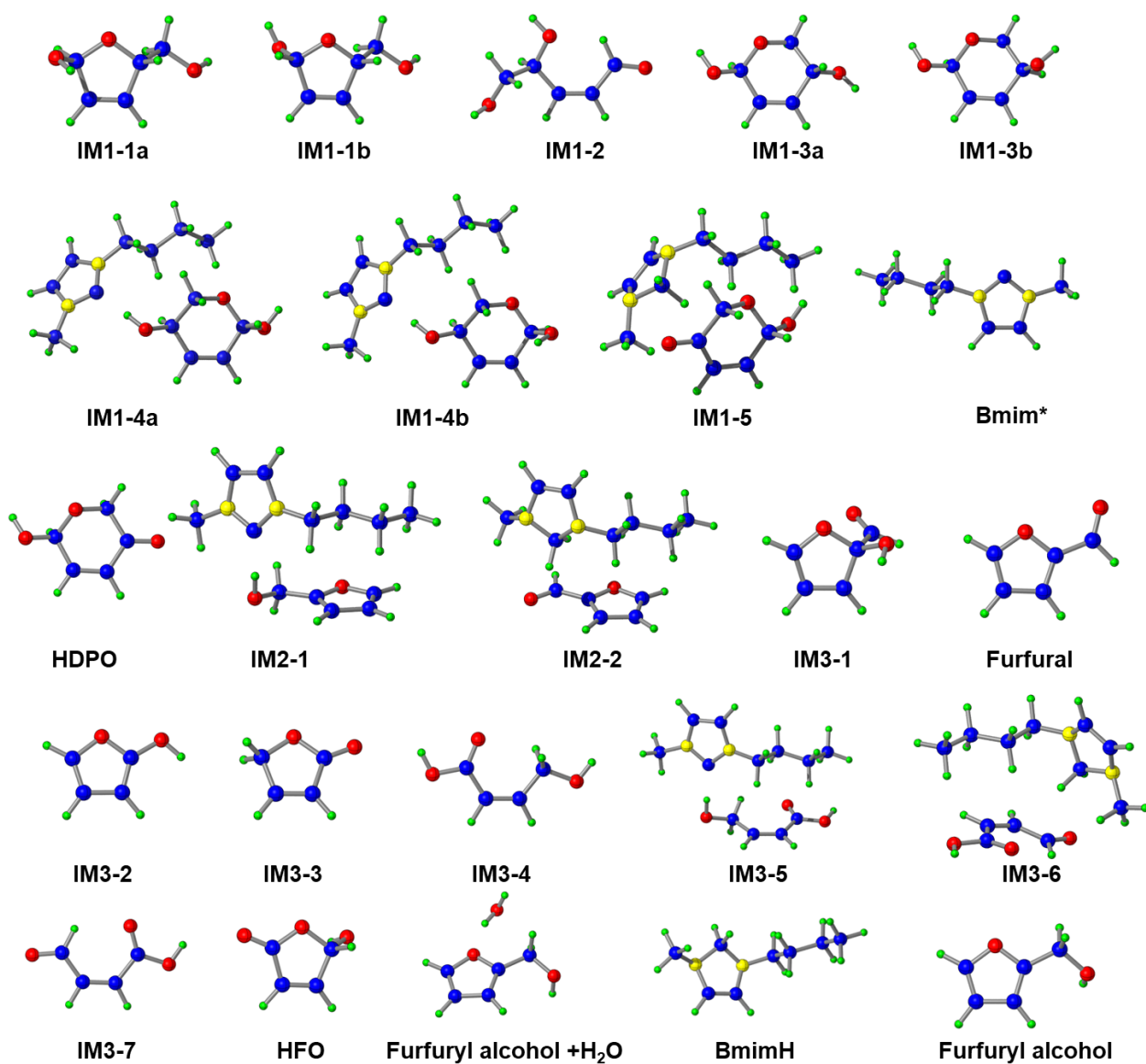


Figure S16. DFT Optimized geometries of the species involved in the catalytic process.

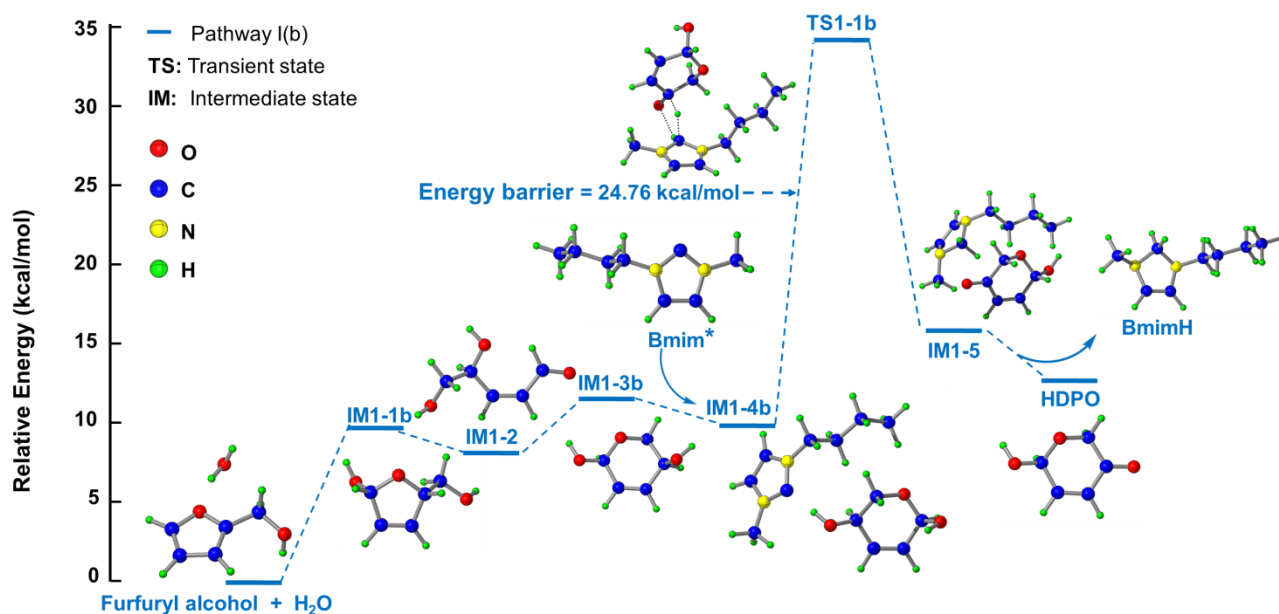


Figure S17. DFT calculation result for the electrochemical oxidation of furfuryl alcohol to HDPO in the ternary electrolyte.

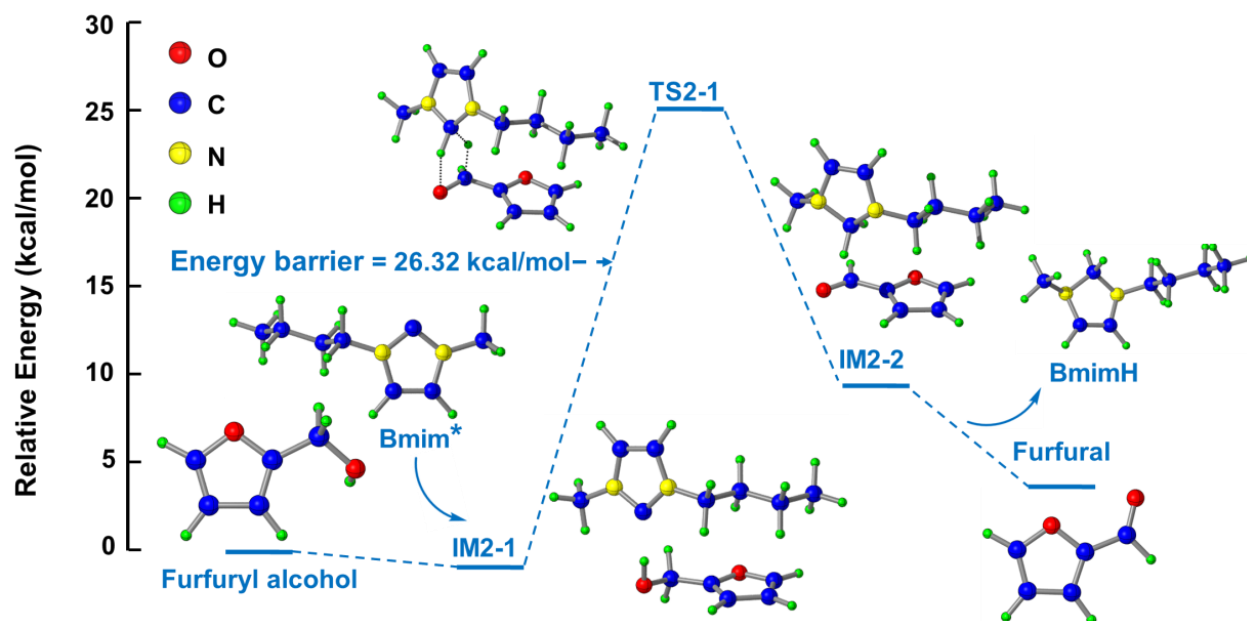


Figure S18. DFT calculation result for the electrochemical oxidation of furfuryl alcohol to furfural in the ternary electrolyte.

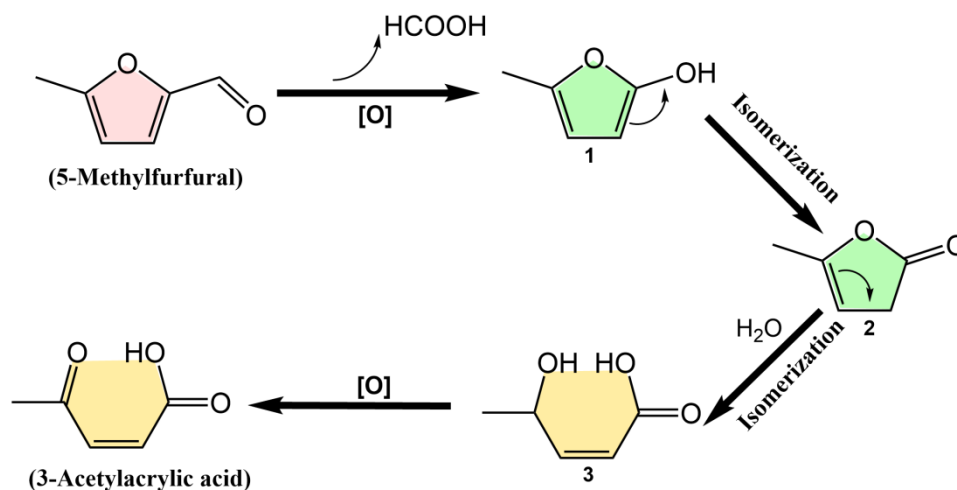


Figure S19. The reaction pathway for the electrochemical oxidation of 5-methylfurfural over the meso-PA/PmPD/GO electrocatalyst in the ternary electrolyte.

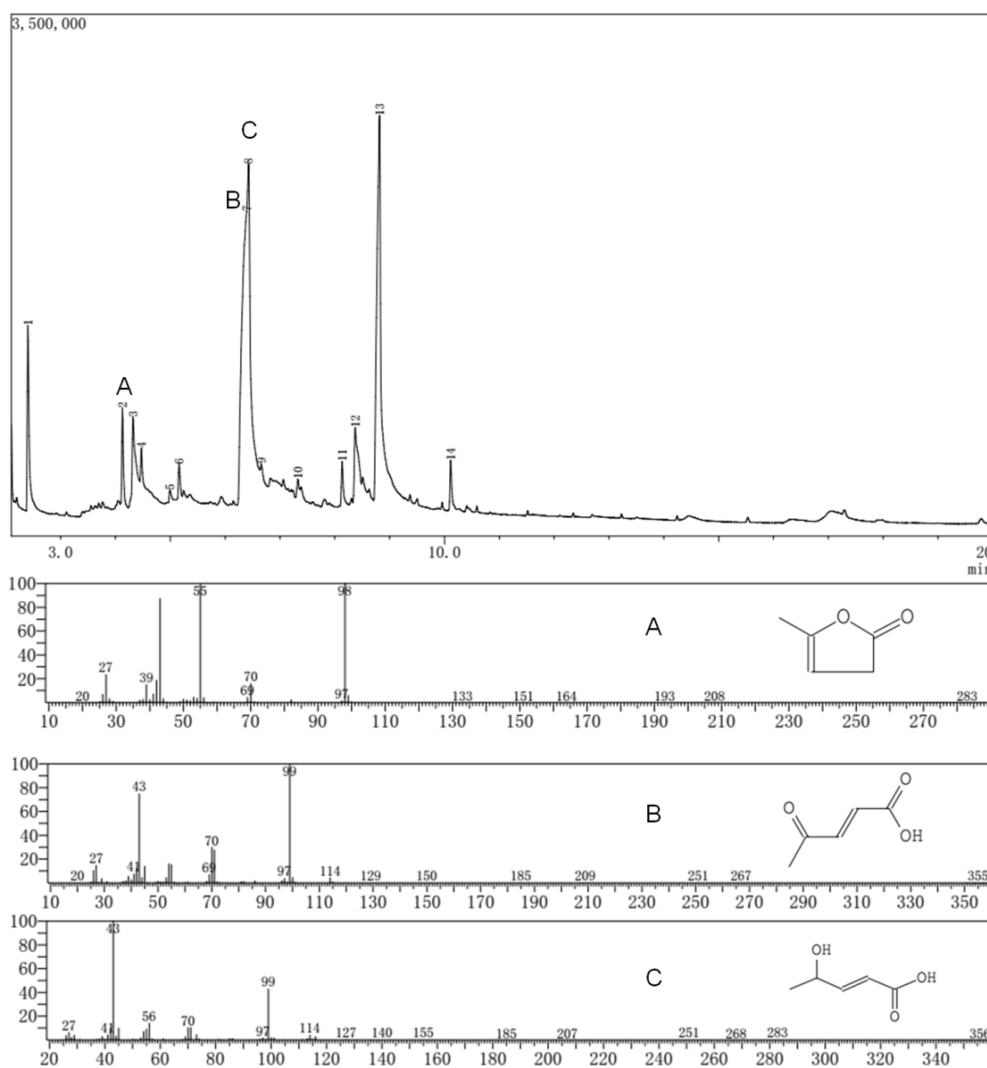


Figure S20. GC-MS spectra of the electrolyte after the electrochemical oxidation of 5-methylfurfural over the meso-PA/PmPD/GO electrocatalyst in the ternary electrolyte.

As shown in Figures S19 and S20, intermediates **2**, **3**, and 3-acetylacrylic acid were detected during the oxidation of 5-methylfurfural on the basis of GC-MS analyses. The results indicated that the aldehyde group in 5-methylfurfural could cleave to generate an intermediate **1** and HCOOH. Then, the unstable **1** rearranged to **2**. Afterwards, compound **3** was formed by ring opening and isomerization of **2**. Finally, 3-acetylacrylic acid was formed *via* the oxidation of **3**.

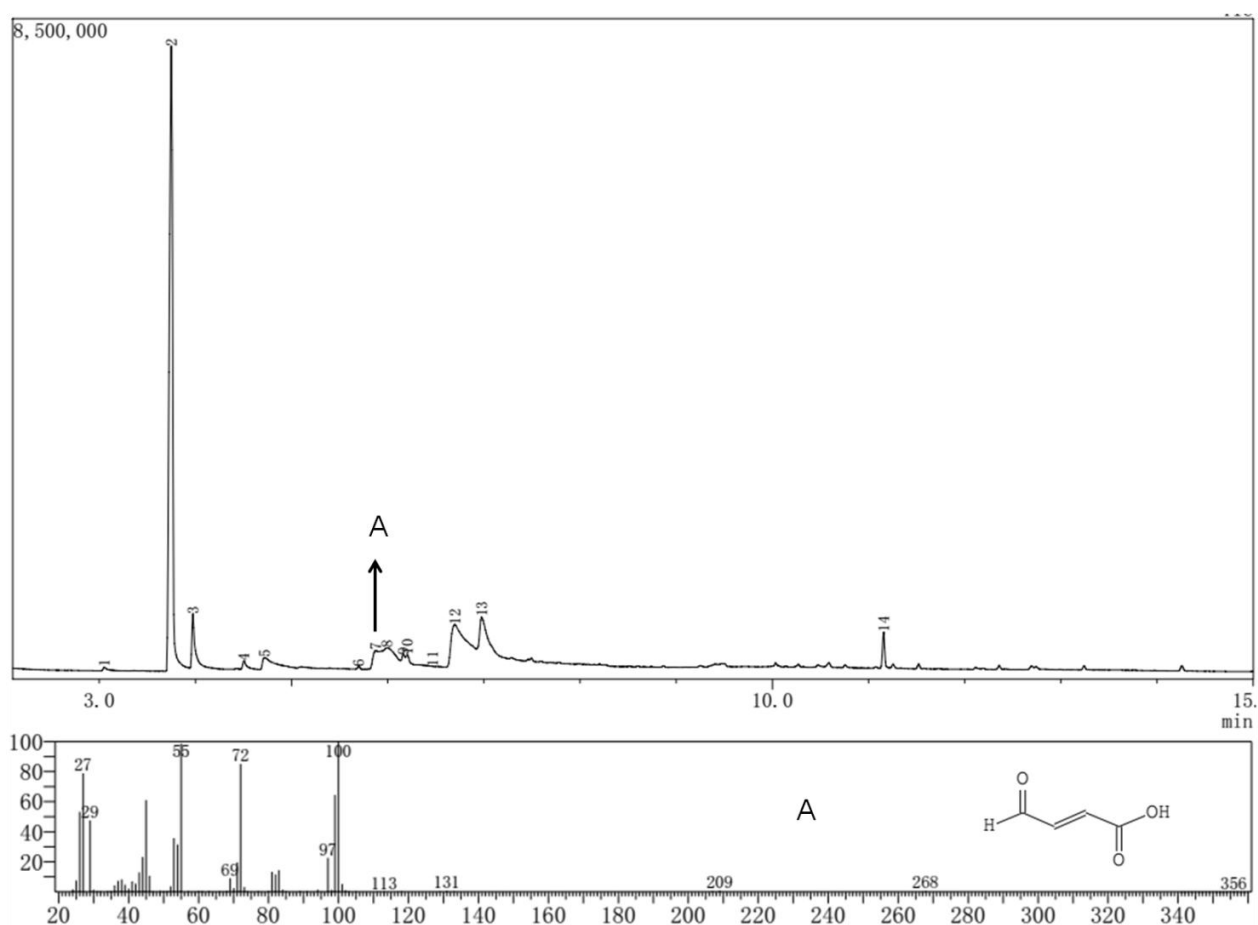


Figure S21. GC-MS spectra of the electrolyte after the electrochemical oxidation of furfural over the meso-PA/PmPD/GO electrocatalyst in the ternary electrolyte.

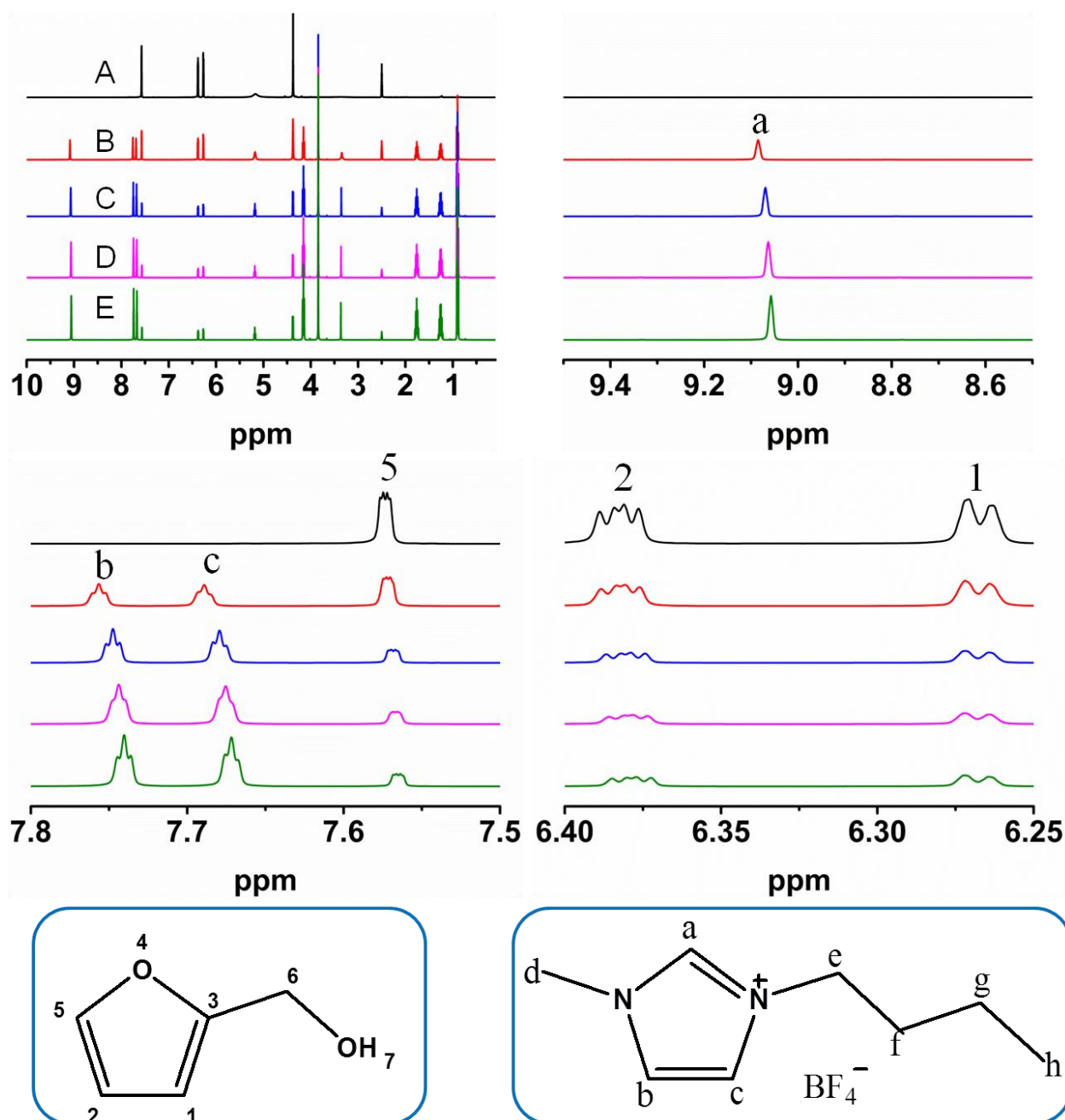


Figure S22. ^1H NMR spectra of the mixture of BmimBF₄ and furfuryl alcohol. (The amount of furfuryl alcohol was 7 μL in all samples, and the amount of BmimBF₄ was 0, 7 μL , 21 μL , 28 μL , and 35 μL in A, B, C, D, and E, respectively. DMSO-*d*₆ was used as solvent).

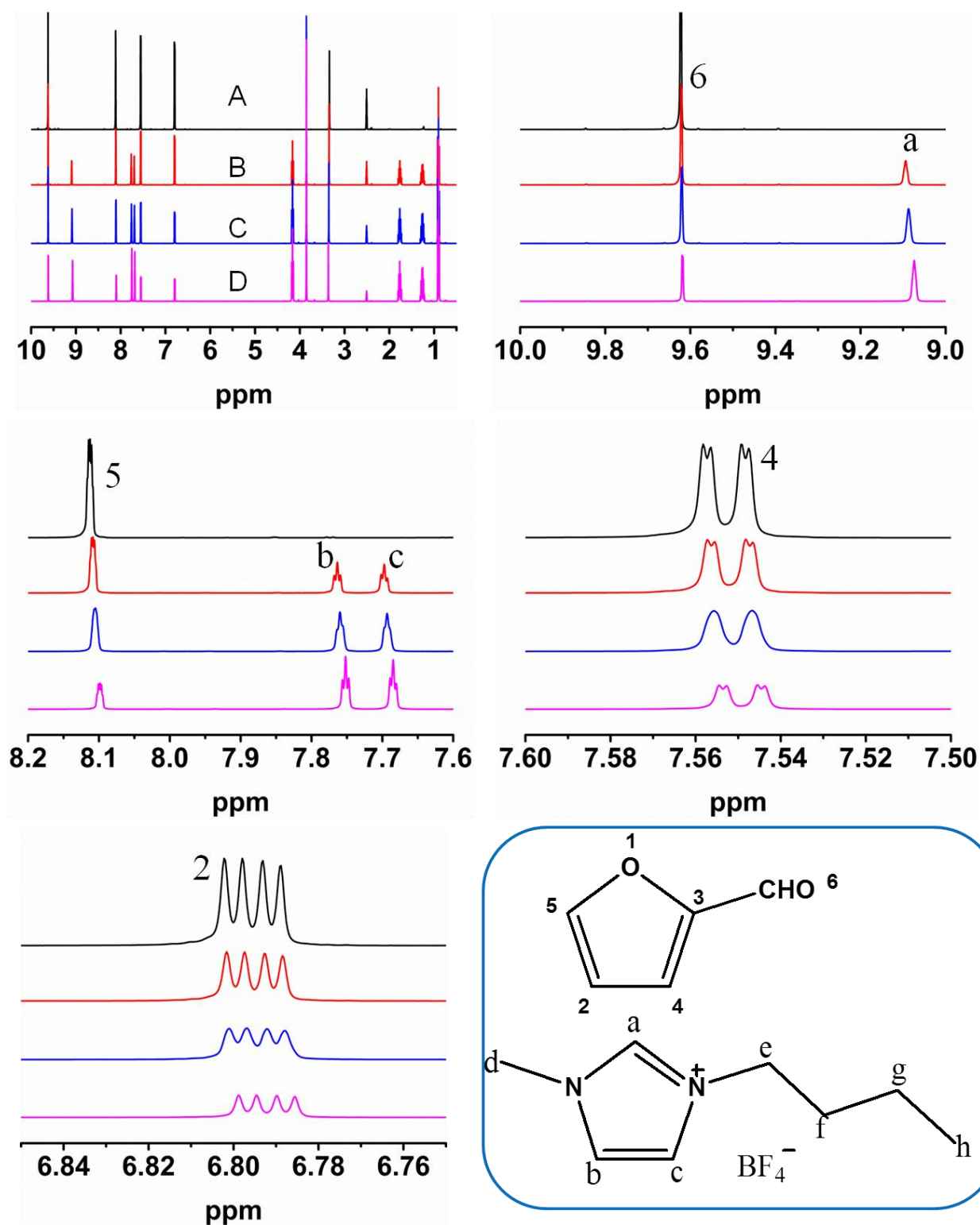


Figure S23. ^1H NMR spectra of the mixture of BmimBF₄ and furfural. (The amount of furfural was 7 μL in all samples, and the amount of BmimBF₄ was 0, 7 μL , 14 μL , and 28 μL in A, B, C, and D, respectively. DMSO-*d*₆ was used as solvent).

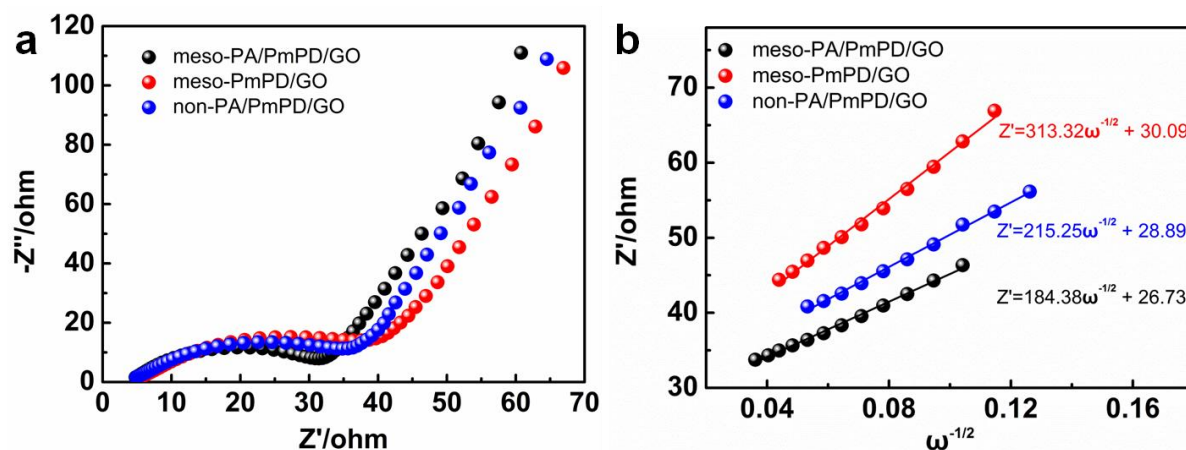


Figure S24. Investigation of charge resistance and mass transport in meso-PA/PmPD/GO and control samples for furfural. (a) EIS spectra. (b) Variations and fittings between Z' and the reciprocal square root of the angular frequency (ω) in the low frequency region of meso-PA/PmPD/GO, meso-PmPD/GO, and non-PA/PmPD/GO in the mixture of furfural and the ternary electrolyte, respectively.

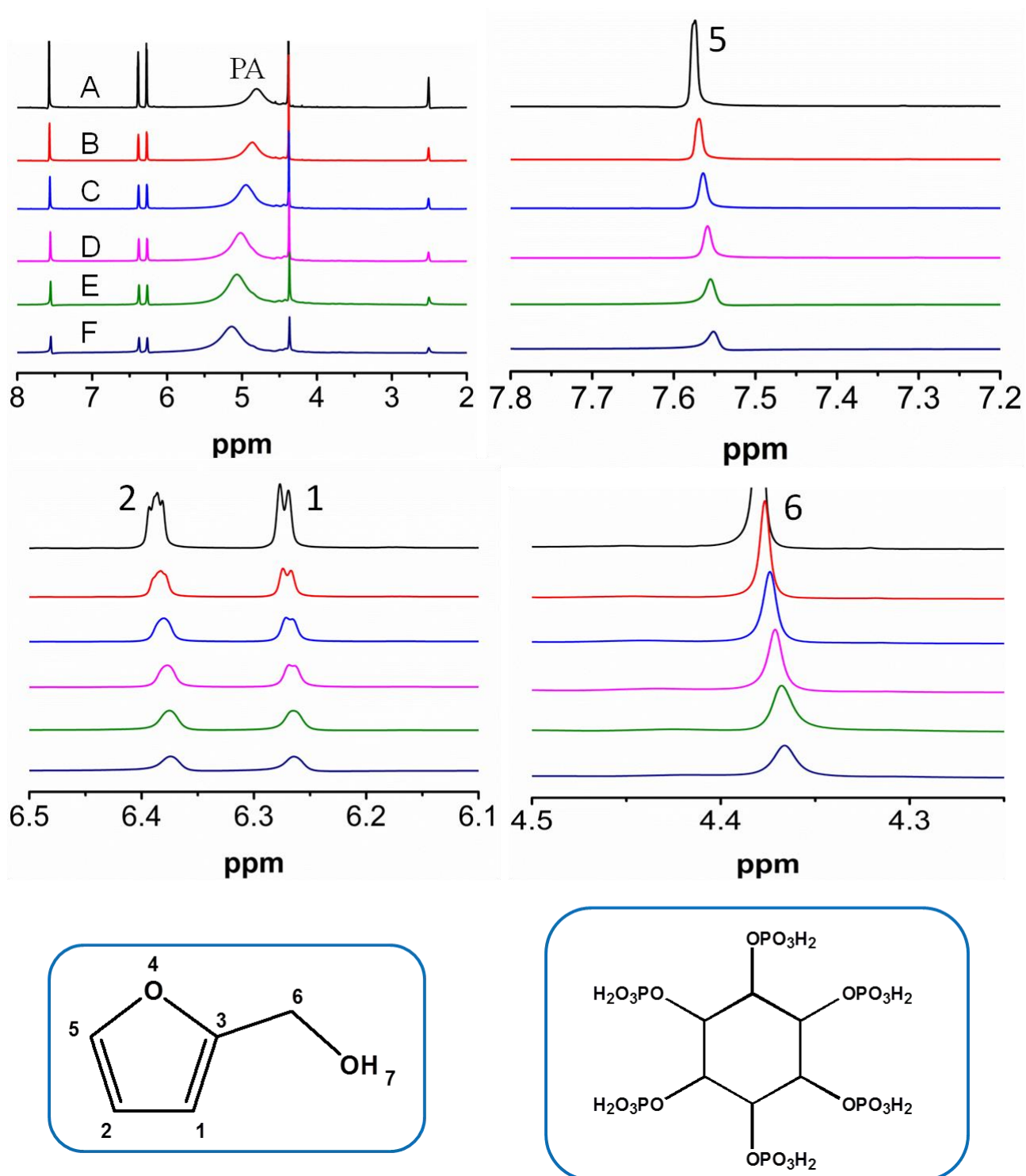


Figure S25. ^1H NMR spectra of the mixture of furfuryl alcohol and PA. (The amount of furfuryl alcohol was 7 μL in all samples, and the amount of PA was 7 μL , 14 μL , 21 μL , 28 μL , 35 μL , and 42 μL in A, B, C, D, E, and F, respectively. $\text{DMSO}-d_6$ was used as solvent).

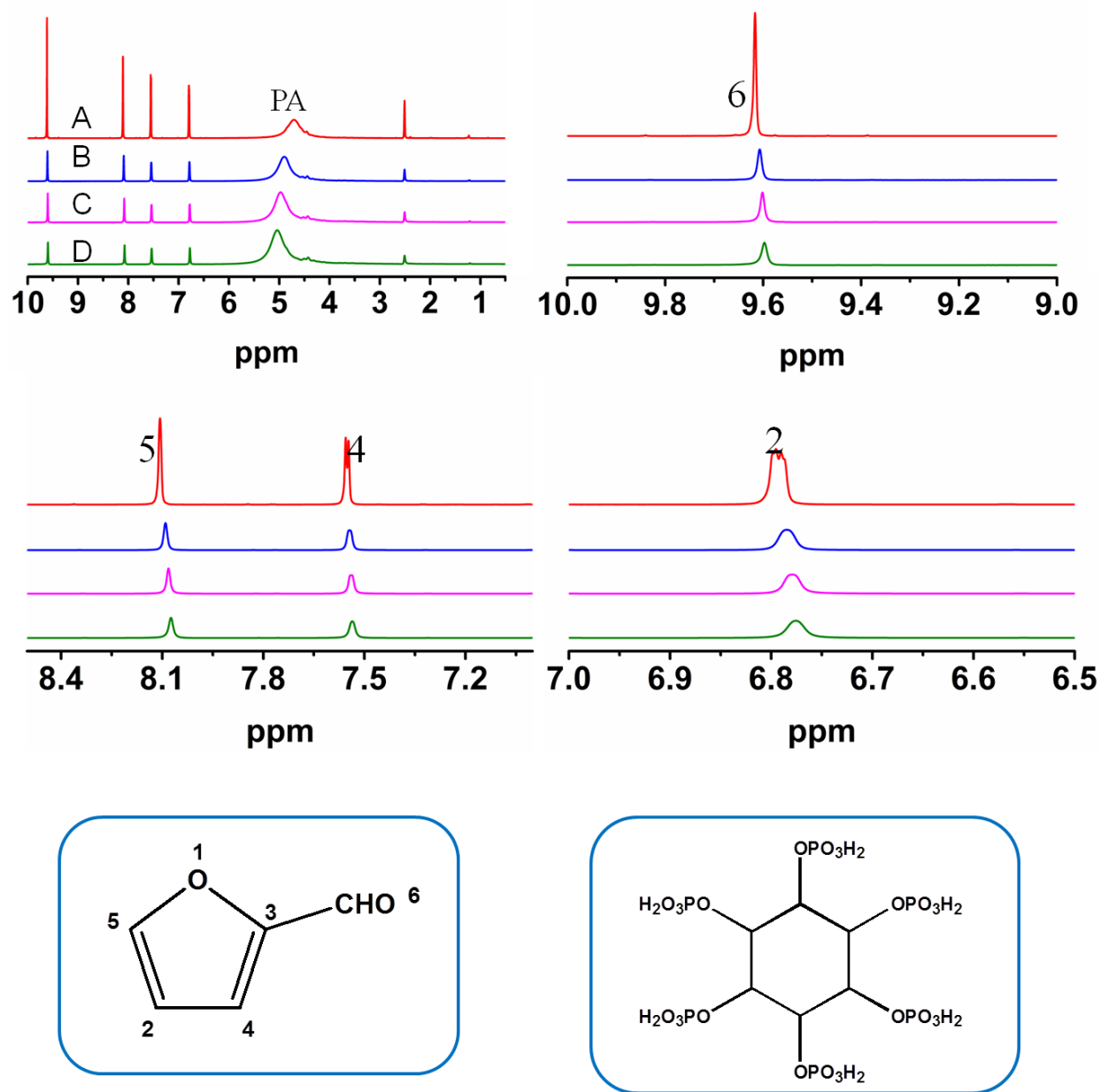


Figure S26. ^1H NMR spectra of the mixture of furfural and PA. (The amount of furfural was 7 μL in all samples, and the amount of PA was 7 μL , 21 μL , 28 μL , and 35 μL in A, B, C, and D, respectively. DMSO- d_6 was used as solvent).

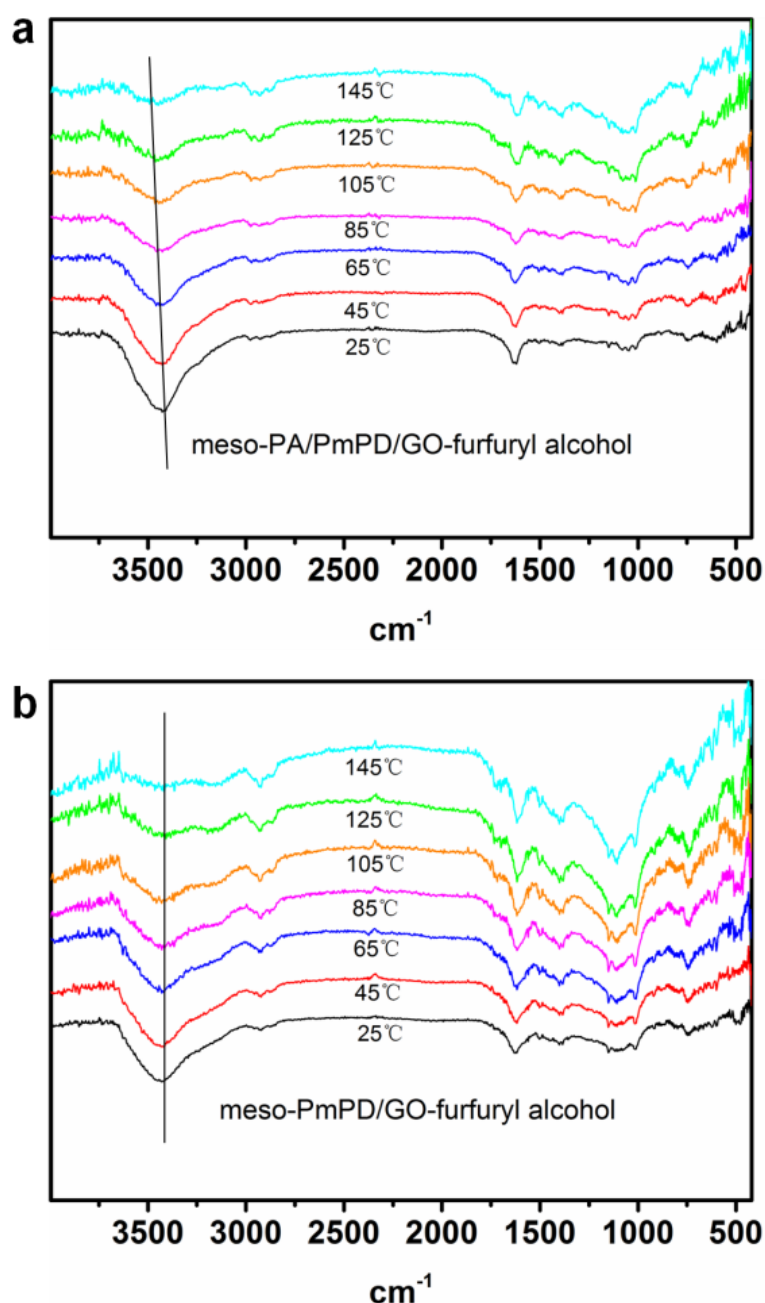


Figure S27. Variable-temperature FTIR spectra of the mixtures of furfuryl alcohol and the 2D catalyst with/without PA. (a) meso-PA/PmPD/GO. (b) meso-PmPD/GO.

As shown in Figure S27, with increasing temperature from 25 to 145 $^{\circ}\text{C}$, the peak attributed to the hydroxyl group (in the range 3200-3500 cm^{-1}) gradually shows a blue shift. This result indicates the existence of H-bonds between meso-PA/PmPD/GO and furfuryl alcohol;¹³⁻¹⁵ in contrast, the H-bonding between the catalyst without PA and furfuryl alcohol could not be detected obviously under the similar condition.

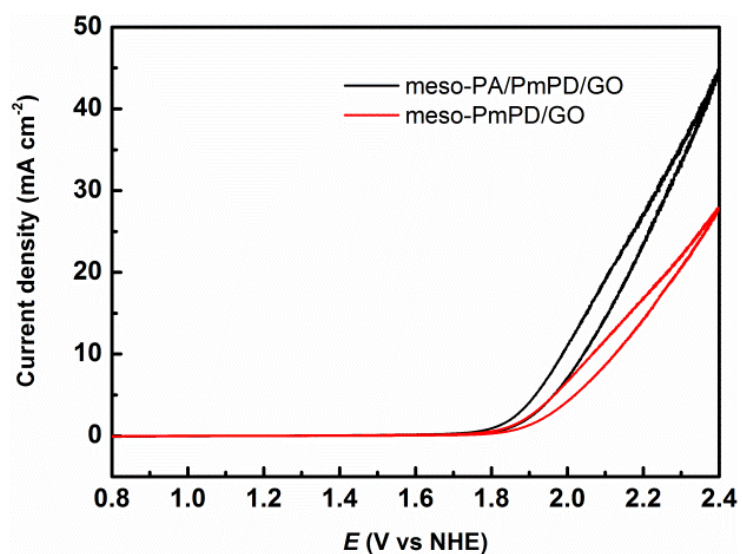


Figure S28. CV curves of the electrocatalytic oxidations of furfuryl alcohol over meso-PA/PmPD/GO and meso-PmPD/GO.

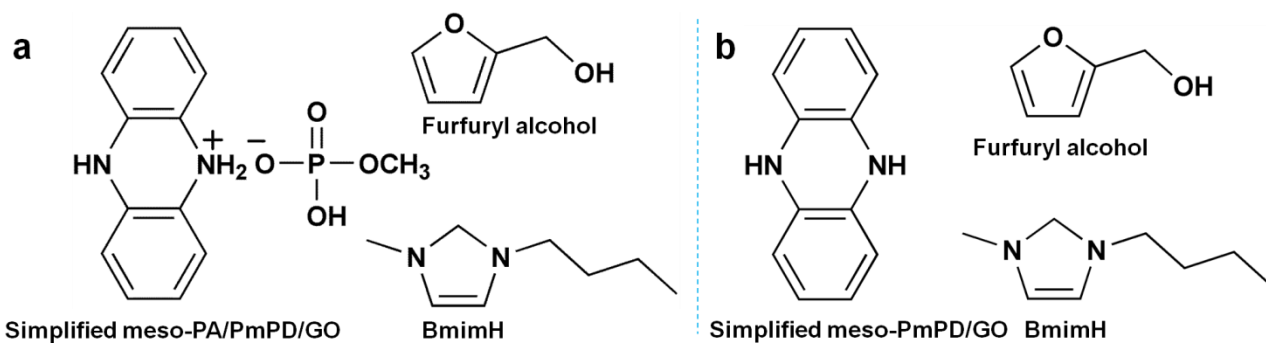


Figure S29. Simplified surface fragments of the 2D catalysts with furfuryl alcohol and BmimH. (a) meso-PA/PmPD/GO with furfuryl alcohol and BmimH. (b) meso-PmPD/GO with furfuryl alcohol and BmimH.

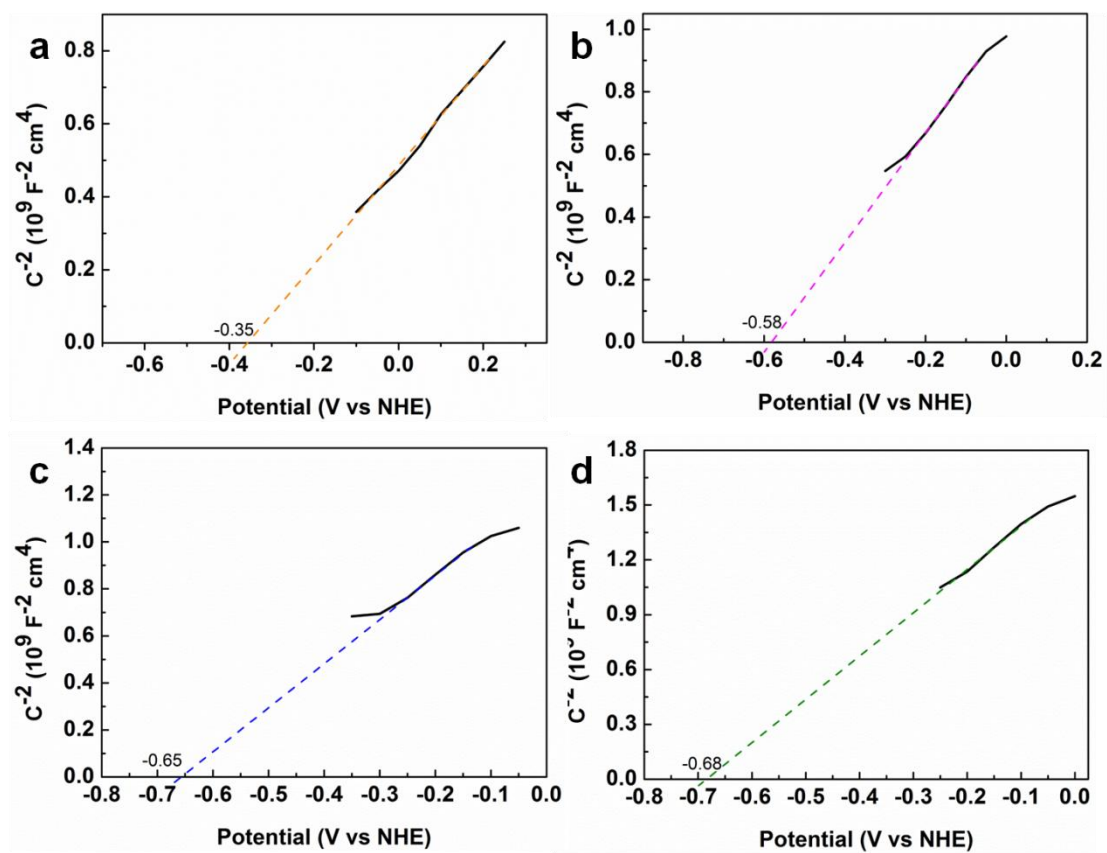


Figure S30. Mott-Schottky curves. (a) meso-PA/PmPD/GO. (b) meso-PmPD/GO. (c) PA/PmPD. (d) PmPD.

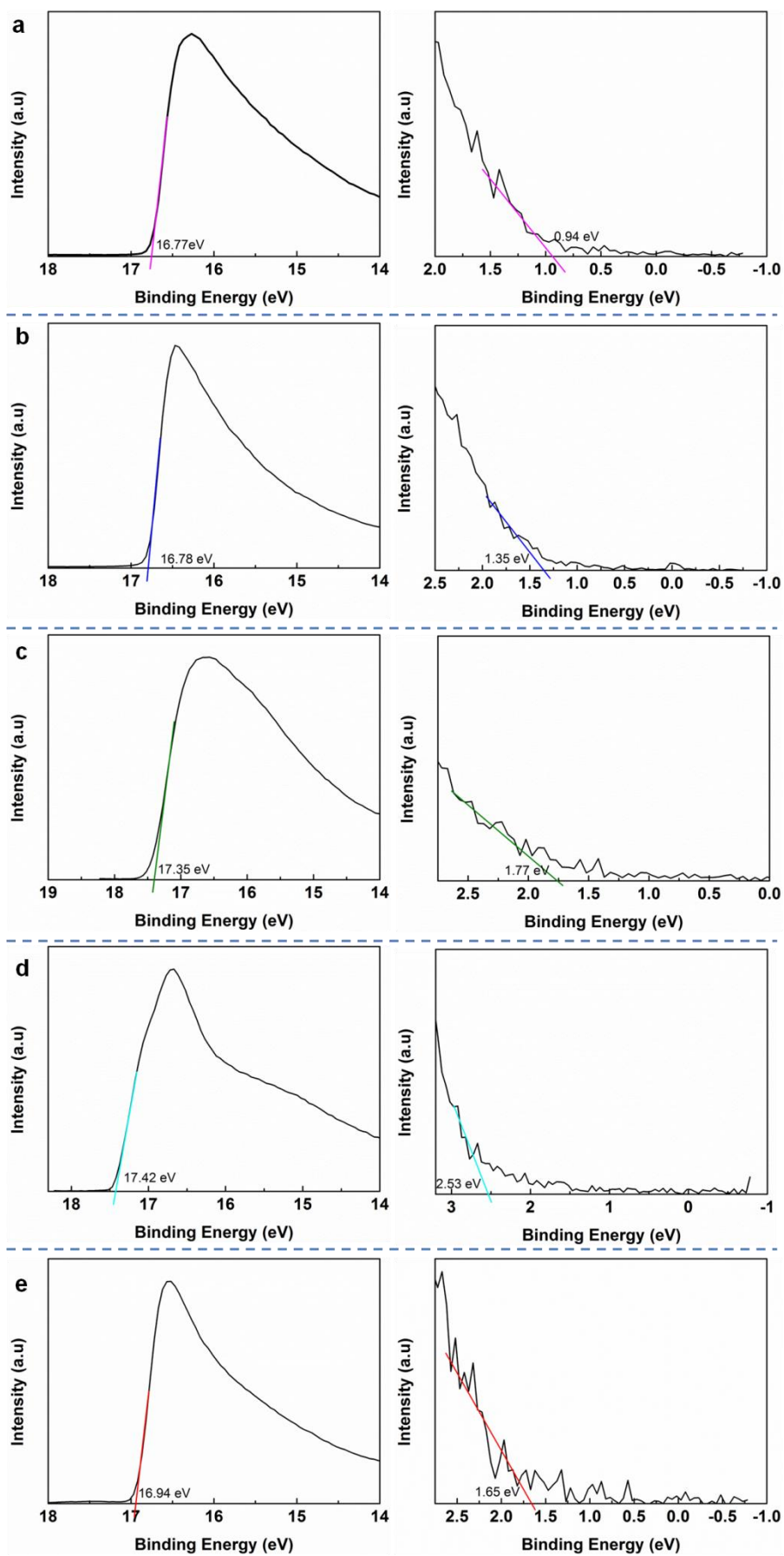


Figure S31. UPS spectra. (a) meso-PA/PmPD/GO. (b) meso-PmPD/GO. (c) PA/PmPD. (d) PmPD. (e) GO.

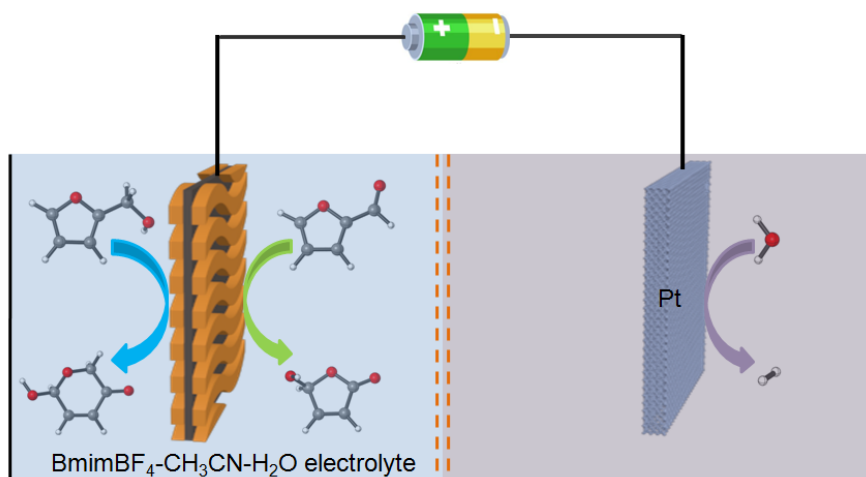


Figure S32. Schematic illustration of an H-type cell for the electrocatalytic oxidation of furan-based molecules. In the anode, the electrons eventually transfer to the cathode through the external circuit.

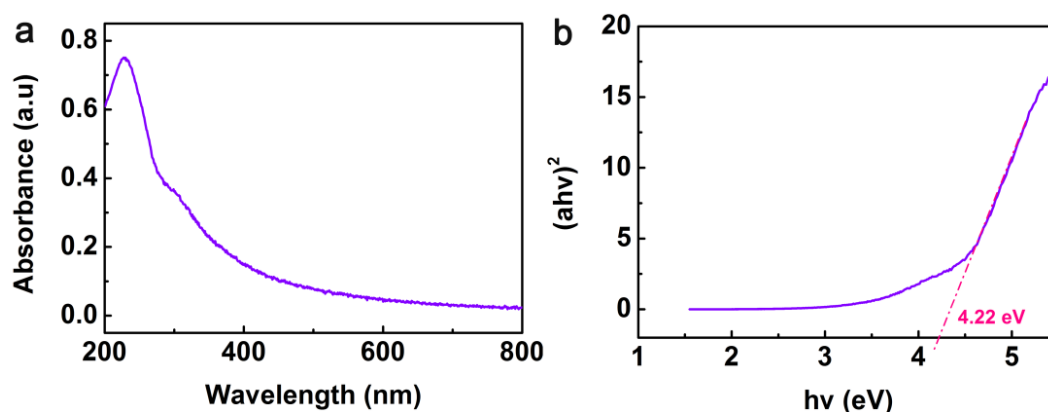


Figure S33. UV-Vis absorption spectra. (a) GO. (b) The corresponding Tauc plot.

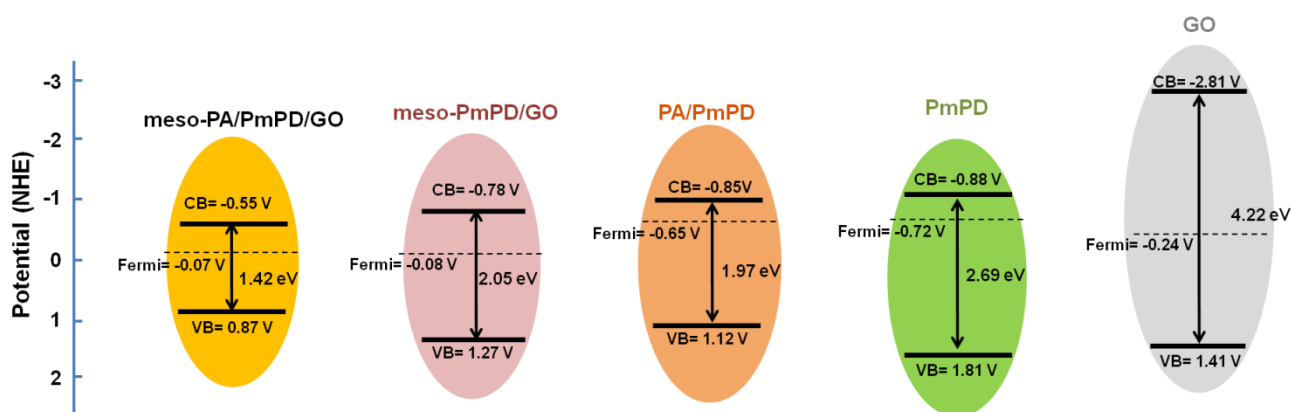


Figure S34. Energy diagrams of meso-PA/PmPD/GO, meso-PmPD/GO, PA/PmPD, PmPD and GO, which are calculated based on Figures S30, S31, and S33.

Table S1. Electrochemical oxidation of furfuryl alcohol over the meso-PA/PmPD/GO electrocatalyst in some inorganic salt (3.4 wt%)-MeCN-H₂O (5.1 wt%) electrolytes.

Electrolytes	Potential (V vs NHE)	Charge (C)	Con. (%)	FE _{HDPO} %	Sel. _{HDPO} %	FE _{furfural} %	Sel. _{furfural} %
KH ₂ PO ₄	2.4	19.5±0.6	10.0±0.6	28.5±1.8	63.9±1.2	6.8±0.7	15.2±0.4
	2.3	13.8±1.9	5.9±1.5	22.4±2.5	61.0±1.4	6.0±1.1	16.2±0.9
NaNO ₃	2.4	83.5±3.5	84.5±0.8	13.5±0.9	15.4±0.8	13.0±1.1	14.8±0.9
	2.3	67.7±2.1	68.9±1.4	14.3±0.7	16.2±0.7	13.4±0.6	15.1±0.8
(NH ₄) ₂ SO ₄	2.4	17.4±0.7	9.1±0.4	28.9±0.7	63.6±1.3	7.2±0.4	15.8±0.9
	2.3	9.5±0.6	4.2±0.6	25.6±3.0	67.5±1.1	6.2±0.9	16.4±0.7

Table S2. Electrochemical oxidation of furfuryl alcohol over the meso-PA/PmPD/GO electrocatalyst in different aqueous inorganic salt electrolytes.

Electrolytes	Potential (V vs NHE)	Charge (C)	Con. (%)	FE _{HDPO} %	Sel. _{HDPO} %	FE _{furfural} %	Sel. _{furfural} %
0.5M KH ₂ PO ₄	1.7	27.2±1.1	7.4±0.9	<2	<2	3.2±0.4	12.0±0.6
	1.8	77.8±1.1	26.0±1.1	18.6±0.8	57.6±1.0	3.3±0.2	10.2±0.4
	1.9	89.2±1.0	34.0±0.5	19.2±0.3	52.1±0.8	7.9±0.4	21.5±1.1
0.5M NaNO ₃	1.7	51.7±1.0	26.9±0.6	15.7±0.3	31.2±0.6	14.7±0.4	29.2±0.7
	1.8	81.5±1.1	41.9±0.9	24.6±0.8	49.6±0.9	9.1±0.3	18.3±0.4
	1.9	93.4±0.8	54.9±1.2	23.5±0.8	41.5±0.9	4.9±0.2	8.8±0.4
0.5M (NH ₄) ₂ SO ₄	1.7	26.3±0.9	15.7±0.6	7.1±0.3	12.3±0.5	18.2±0.7	31.7±0.9
	1.8	60±0.6	38.6±0.9	23.6±0.6	38.1±0.4	14.1±0.5	22.7±0.8
	1.9	84.7±0.6	51.7±0.8	42.7±0.7	72.6±0.7	3.2±0.4	5.4±0.7

Table S3. The effect of ionic liquids on electrochemical oxidations of furfuryl alcohol at 2.3 V vs NHE in the ternary electrolyte of IL (3.4 wt%)-MeCN-H₂O (5.1 wt%).

IL	Charge (C)	Con. (%)	FE _{HDPO} %	Sel. _{HDPO} %	FE _{furfural} %	Sel. _{furfural} %
BmimBF ₄	88.9±1.7	97.7±1.5	82.2±1.3	86.1±1.6	6.6±0.4	6.9±0.4
BmimPF ₆	91.1±1.4	96.9±1.6	72.0±1.7	77.9±1.6	10.9±1.7	11.7±1.5
BmimCH ₃ COO	89.8±1.3	83.4±2.2	69.8±1.5	86.5±1.9	7.6±1.0	6.2±0.9
BmimNO ₃	92.2±0.6	98.1±0.7	74.6±0.5	80.7±0.5	12.0±0.6	13.0±0.7
BmimH ₂ PO ₄	92.3±0.9	97.7±1.6	72.9±1.0	79.4±0.8	15.1±0.7	16.4±0.8

Table S4. The effect of PA content in meso-PA/PmPD/GO on electrochemical oxidations of furfuryl alcohol at 2.3 V vs NHE in the ternary electrolyte of BmimBF₄ (3.4 wt%)-MeCN-H₂O (5.1 wt%).

PA content	Charge (C)	Con. (%)	FE _{HDPO} %	Sel. _{HDPO} %	FE _{furfural} %	Sel. _{furfural} %
0	85.5±1.5	85.4±0.9	66.9±0.5	77.2±1.1	12.6±0.7	14.5±0.7
5.3	94.5±1.0	92.3±2.1	65.0±0.7	76.6±0.7	8.9±0.4	10.5±0.5
12.5	93.6±1.2	94.2±1.4	70.4±2.0	80.5±0.8	8.2±0.7	9.4±0.6
13.1	88.9±1.7	97.7±1.5	82.2±1.3	86.1±1.6	6.6±0.4	6.9±0.4
19.1	98.6±2.7	96.4±1.9	61.6±1.3	72.5±2.2	7.8±0.4	9.3±0.4

Table S5. Electrochemical oxidation of furfural to HFO over the meso-PA/PmPD/GO electrocatalyst in different aqueous inorganic salt electrolytes.

Electrolytes	Potential (V vs NHE)	Charge (C)	Con. (%)	FE _{HFO} %	Sel. _{HFO} %
0.5M KH ₂ PO ₄	1.8	48.1±0.8	9.8±0.5	27.4±1.5	70.0±0.5
	1.9	70.5±1.1	25.9±0.6	49.1±0.6	69.1±0.7
	2.0	89.4±0.9	36.8±0.4	58.7±1.1	73.9±0.7
0.5M NaNO ₃	1.8	67.7±1.1	20.7±0.6	32.7±0.8	55.5±0.8
	1.9	90.4±1.2	36.5±1.0	51.3±0.5	65.8±1.3
	2.0	126.8±1.2	43.1±0.9	42.8±0.9	65.2±0.7
0.5M (NH ₄) ₂ SO ₄	1.8	67.4±1.5	10.6±1.3	15.5±1.5	51.2±0.6
	1.9	82.1±1.5	18.5±1.1	27.2±1.6	62.7±1.0
	2.0	109.3±0.7	29.4±0.8	36.0±1.1	69.2±0.5

Table S6. DFT calculated energy profile.

Intermediate State	Single Point Energy	Gibbs Free Energy
Furfuryl alcohol + H ₂ O	-421.136418	-421.046001
IM1-1a	-421.128247	-421.031391
IM1-1b	-421.127003	-421.030323
IM1-2	-421.123889	-421.031692
IM1-3a	-421.126947	-421.02852
IM1-3b	-421.125986	-421.027533
IM1-4a	-844.055096	-843.761474
IM1-4b	-844.057709	-843.765006
TS1-1a	-844.012199	-843.723256
TS1-1b	-844.014765	-843.725542
IM1-5	-844.043079	-843.752528
HDPO	-419.923451	-419.847826
Furfuryl alcohol	-344.671873	-344.599762
IM2-1	-767.601192	-767.33564
TS2-1	-767.556722	-767.293702
IM2-2	-767.583776	-767.319738
Furfural	-343.465203	-343.415199
IM3-1	-419.272354	-419.212374
TS3-1	-419.234936	-419.178195
IM3-2	-305.346627	-305.300785
IM3-3	-305.374982	-305.328473
IM3-4	-381.833487	-381.766637
IM3-5	-804.763907	-804.501865
TS3-2	-804.717931	-804.458932
IM3-6	-804.738402	-804.478403
IM3-7	-380.619352	-380.574498
HFO	-380.623367	-380.573773

Table S7. The effect of meso-PA/PmPD/GO amount on electrochemical oxidations of furfuryl alcohol at 2.3 V vs NHE in the ternary electrolyte of BmimBF₄ (3.4 wt%)-MeCN-H₂O (5.1 wt%).

meso-PA/PmPD/GO amount (mg)	Charge (C)	Con. (%)	FE _{HDPO} %	Sel. _{HDPO} %	FE _{furfural} %	Sel. _{furfural} %
10	88.9±1.7	97.7±1.5	82.2±1.3	86.1±1.6	6.6±0.4	6.9±0.4
12	89.9±1.4	98.3±1.1	83.3±0.8	87.7±1.1	8.0±0.7	8.5±0.7
15	89.4±1.3	98.1±0.7	82.8±1.5	87.0±1.5	6.8±0.4	7.1±0.4

5. Coordinates of DFT-computed stationary points

Furfuryl alcohol+H₂O:

C	-0.98810400	1.16230800	0.22914500
H	-1.34520400	1.14424100	1.26361100
C	0.27647100	0.37372100	0.10889000
C	1.58377200	0.72175400	-0.01198600
O	0.16165000	-0.99394600	0.11584300
C	2.33077400	-0.50577200	-0.08410400
H	1.97196200	1.72700000	-0.04116900
C	1.42907700	-1.51294400	-0.00056500
H	3.39958500	-0.61362600	-0.18500200
H	1.49917500	-2.58778000	-0.00772000
H	-1.76686400	0.69677600	-0.38659500
O	-0.78397900	2.52981000	-0.09734600
H	-0.52534900	2.56558300	-1.02711400
O	-2.67131600	-1.69485400	-0.21657500
H	-2.96782200	-1.41457800	0.65749600
H	-1.70826500	-1.68010800	-0.13716600

IM1-1a:

C	1.84834200	-0.58315800	0.33977800
H	2.02020000	-1.65941300	0.21524700
C	0.59436000	-0.19337900	-0.43437200
C	0.14374000	1.21721300	-0.19219700
O	-0.49402800	-1.00880100	0.03072400
C	-1.06786200	1.21657400	0.35115700
H	0.75641300	2.07461100	-0.43474300
C	-1.58959200	-0.19121300	0.45099700
H	-1.66123900	2.07102800	0.64815000
H	1.68705400	-0.37662500	1.40567700
O	2.92770900	0.18371100	-0.19004500
H	3.72301100	-0.05767100	0.29739000
H	0.77478600	-0.37995000	-1.50100000
O	-2.73848800	-0.41414100	-0.33629200
H	-1.88335200	-0.49550700	1.45866000
H	-2.55233900	-0.05885500	-1.21666600

IM1-1b:

C	-1.58137900	-0.73733500	-0.16955100
H	-1.77521300	-1.67381000	0.36812000
C	-0.63643600	0.12084600	0.66695000
C	-0.15429900	1.35486900	-0.04030400
O	0.56284200	-0.61611900	0.95997200
C	1.15317900	1.27051800	-0.25283900
H	-0.81752000	2.15526800	-0.33807200
C	1.66525500	-0.04772400	0.25366900
H	1.78972900	1.98169800	-0.76134800
H	-1.09245900	-0.97713600	-1.12194800
O	-2.77929800	0.01297800	-0.36233200
H	-3.37565800	-0.52277100	-0.89683800
H	-1.12803000	0.36109600	1.61646500
H	2.49773400	0.02851100	0.96169100
O	2.05583700	-0.83499700	-0.85583700
H	2.50845000	-1.61480500	-0.51004300

IM1-2:

C	-2.13807700	0.27047400	0.45287500
H	-2.70751900	1.20576500	0.37017800
C	-0.87284000	0.40735600	-0.40734000
C	-0.01325300	-0.80552000	-0.26996900
O	-0.18003900	1.59735300	-0.05110200
C	1.30773800	-0.93877100	-0.06882600
H	-0.59907400	-1.71687000	-0.34975500
C	2.31709900	0.12154800	0.04404400
H	1.71660000	-1.94318000	0.01098300
H	-1.84463600	0.13423100	1.50051500
O	-2.89278000	-0.83373300	-0.03661400
H	-3.58151000	-1.02787400	0.60891100
H	-1.20037100	0.44803600	-1.45810600
H	1.96605600	1.15762500	-0.05896100
O	3.49623700	-0.13020100	0.23352000
H	-0.74090100	2.34439700	-0.29489900

IM1-3a:

C	0.62551500	-1.20431800	0.16414100
C	0.64932900	1.25212400	-0.15906400
C	-0.63039700	1.25696900	0.20424200
C	-1.40978400	-0.01800700	0.36515900
H	1.20054500	2.18129500	-0.27864900
H	0.72092500	-1.18710400	1.25888500
H	-1.16576200	2.17518500	0.42241000
O	-0.74087300	-1.12737100	-0.23025400
H	1.01201800	-2.15410500	-0.20540900
H	-1.54745600	-0.23738200	1.43851500
O	-2.64806900	0.13477000	-0.26483700
H	-3.17832200	-0.64200600	-0.04458600
C	1.40776900	-0.02657500	-0.40717100
O	2.67744200	-0.02403300	0.25703400
H	1.54949800	-0.16841300	-1.48694500
H	3.24596100	0.60444700	-0.20361000

IM1-3b:

C	0.65813600	-1.13074700	0.61388000
C	0.65926600	1.33100900	0.25252300
C	-0.66868300	1.26342800	0.23837900
C	-1.39725300	-0.05162900	0.26400300
H	1.17254900	2.28626900	0.20833800
H	0.43587900	-1.16161100	1.69110700
H	-1.28953000	2.15323000	0.22482100
O	-0.55448600	-1.12459600	-0.13702100
H	1.19013500	-2.04487200	0.34605000
H	-1.75701700	-0.26019300	1.28823700
O	-2.47193700	0.02220400	-0.62645300
H	-2.98496400	-0.78931400	-0.52116100
C	1.51702900	0.09733600	0.31414100
O	2.20836800	0.00112900	-0.94155600
H	2.24575100	0.20910600	1.12855500
H	2.92066600	-0.63889400	-0.82326400

IM1-4a:

C	1.68892300	-0.40546200	1.11909000
C	1.94565100	-2.27092700	-0.47199800
C	3.24104500	-1.96916900	-0.53156300
C	3.79513600	-0.70418500	0.06356200
H	1.56090000	-3.19229200	-0.90003500
H	2.04807700	-0.97128600	1.99013700
H	3.96762200	-2.63309700	-0.98861800
O	2.77573300	0.21813000	0.43367400
H	1.03878500	0.39501100	1.46966800
H	4.38137000	-0.94667200	0.96730800
O	4.61339300	-0.08368800	-0.88983700
H	5.05311600	0.65457600	-0.44851100
C	0.94414800	-1.36643900	0.19116500
O	-0.01074600	-2.12104500	0.91483200
C	-4.28741800	-0.11369100	-0.71921700
C	-3.73246200	1.06670700	-0.34700800
C	-2.36359400	-0.59295500	0.40703900
N	-3.43527900	-1.10481800	-0.25354500
H	-5.19442100	-0.33277100	-1.25763400
H	-4.05576700	2.08228700	-0.50351000
N	-2.56880200	0.75052200	0.33927600
C	-1.61474200	1.74510100	0.83206300
H	-2.18054200	2.60429100	1.19927100
H	-1.09262100	1.29934200	1.67902300
C	-3.66921400	-2.53153500	-0.43402000
H	-2.76289200	-3.06319300	-0.15104700
H	-4.49931900	-2.86855600	0.19066500
H	-3.89920300	-2.74170900	-1.47968300
C	-0.61551500	2.17050800	-0.24226400
H	-1.15493900	2.62326900	-1.08168500
H	-0.12221800	1.27455000	-0.62948500
C	0.43858600	3.14393700	0.28743200
H	-0.04136100	4.09137100	0.55887500
H	0.87050800	2.73907300	1.20970500
C	1.55856700	3.39211800	-0.72328200
H	1.16196300	3.78062600	-1.66719100
H	2.28340200	4.11830100	-0.34293400
H	2.09462100	2.46222800	-0.93298200
H	0.43573900	-0.78095000	-0.58876100
H	-0.86594900	-1.60555000	0.85195800

IM1-4b:

C	1.85511000	-0.08286600	0.76803600
C	2.11448000	-2.13127200	-0.57180000
C	3.37090300	-1.75561100	-0.80531600
C	3.87428800	-0.37642100	-0.46782500
H	1.76922400	-3.13189000	-0.81580200
H	2.32486000	-0.49700200	1.66978600
H	4.10204800	-2.43482700	-1.23192600
O	2.83319800	0.50930400	-0.09523600
H	1.18258100	0.71860400	1.06957800
H	4.34258200	0.09360800	-1.33418400
O	4.90081900	-0.44050600	0.51342400
H	4.61780800	-1.07046400	1.18885000
C	1.10679300	-1.20351700	0.04968700
O	0.27459500	-1.90949600	0.94981100
C	-4.29407400	-0.53837900	-0.56733200
C	-3.88370600	0.70537900	-0.21354900
C	-2.26447500	-0.75555700	0.44644600
N	-3.29179600	-1.40673100	-0.15865200
H	-5.19183800	-0.87663600	-1.05727300
H	-4.34991000	1.66813300	-0.34081400
N	-2.65188100	0.54718800	0.40466100
C	-1.81191600	1.65584400	0.85842400
H	-2.46482700	2.43226800	1.26313800
H	-1.19197600	1.27874900	1.67250200
C	-3.33735800	-2.85118500	-0.34325900
H	-2.36949000	-3.25929800	-0.05894800
H	-4.11672600	-3.29420400	0.28036600
H	-3.53712400	-3.08861900	-1.38949800
C	-0.93374400	2.21231400	-0.26021500
H	-1.56918400	2.57924100	-1.07387200
H	-0.33433900	1.39328000	-0.66962500
C	-0.01097300	3.33041400	0.22678800
H	-0.61297900	4.19412200	0.53185200
H	0.52155400	2.99190500	1.12319900
C	1.00633900	3.74911900	-0.83433200
H	0.50843700	4.08420000	-1.75015200
H	1.63823900	4.56865500	-0.47937400
H	1.65903600	2.90947900	-1.09128300
H	0.49856500	-0.76068200	-0.75451800
H	-0.63971500	-1.50582000	0.86743300

TS1-1a:

C	1.01991900	-1.18374300	1.41914900
C	-0.25562900	-2.56168800	-0.23825700
C	0.89783700	-2.76269300	-0.88293600
C	2.16001700	-2.04844500	-0.48930400
H	-1.15932600	-3.09362300	-0.52151200
H	1.43961400	-1.95896400	2.07961300
H	0.98826900	-3.46980400	-1.70151400
O	1.90829700	-0.91523600	0.33466600
H	0.93301100	-0.25938200	1.99175900
H	2.82609600	-2.73431900	0.06346200
O	2.79660600	-1.59265300	-1.65268700
H	3.65722300	-1.24292600	-1.38804600
C	-0.35989400	-1.66053000	0.95323500
O	-1.23434800	-1.88711400	1.86093900
C	-2.98280200	1.21588500	-1.30873200
C	-2.10378300	2.17944400	-0.94870700
C	-1.84550200	0.45630100	0.49330800
N	-2.88889900	0.19280900	-0.37388400
H	-3.68505200	1.17299900	-2.12398600
H	-1.88817000	3.13547900	-1.39421600
N	-1.46449900	1.75521200	0.21342000
C	-0.27498100	2.37694200	0.78412800
H	-0.48305300	3.43903900	0.93659600
H	-0.11665700	1.92517900	1.76556200
C	-3.55898100	-1.09185400	-0.44154800
H	-3.39836600	-1.60951800	0.50363800
H	-4.62599100	-0.94544300	-0.61626600
H	-3.14325900	-1.70593600	-1.24614600
C	0.95800200	2.18079300	-0.10079200
H	0.81695300	2.73484100	-1.03579200
H	1.04729600	1.12241500	-0.35565900
C	2.24833400	2.63303300	0.58198500
H	2.18227500	3.69765400	0.83720400
H	2.35819100	2.09238700	1.52919800
C	3.47940200	2.38273600	-0.28915800
H	3.41436700	2.93438000	-1.23283400
H	4.39863800	2.69393800	0.21601300
H	3.56758900	1.31901900	-0.52820300
H	-0.80053600	-0.48289500	0.28620400
H	-1.91140400	0.06226200	1.52060800

TS1-1b:

C	1.55858900	-0.43092300	1.06310300
C	0.81560500	-2.51045600	-0.10227100
C	1.82203900	-2.32773500	-0.96482600
C	2.65113600	-1.07301000	-0.96907500
H	0.23060800	-3.42563700	-0.10292200
H	2.37382600	-0.82070500	1.69100600
H	2.10216400	-3.08579800	-1.68957600
O	2.06155800	-0.02674900	-0.21634500
H	1.16450400	0.46478700	1.54367300
H	2.74284700	-0.66506200	-1.97712800
O	3.98675100	-1.34202300	-0.55956400
H	3.94117000	-1.95277400	0.18743400
C	0.47705900	-1.50592300	0.95244100
O	-0.11497400	-1.87353400	2.02565400
C	-3.31694300	-0.20558600	-1.19423800
C	-2.91380700	1.05832200	-0.92450100
C	-1.82956600	-0.31694200	0.50719700
N	-2.72165200	-1.04245800	-0.25920200
H	-3.99129100	-0.58469100	-1.94317000
H	-3.17049900	1.99272900	-1.39343400
N	-2.06522800	1.00208200	0.17668700
C	-1.30371100	2.12067100	0.71741800
H	-1.99229400	2.95322600	0.88373200
H	-0.92268500	1.80664200	1.69130500
C	-2.76276200	-2.49131800	-0.25621000
H	-2.44739000	-2.84458100	0.72532700
H	-3.77880200	-2.83009500	-0.46042600
H	-2.08531200	-2.90489500	-1.00952000
C	-0.15075900	2.54712300	-0.19285400
H	-0.56149700	2.93921900	-1.13021700
H	0.45104900	1.67041600	-0.44523700
C	0.73861900	3.60316300	0.46431000
H	0.13897600	4.48698300	0.71278900
H	1.11789500	3.20930000	1.41477900
C	1.91556200	4.00947800	-0.42230500
H	1.56802700	4.42568400	-1.37348400
H	2.54030100	4.76458000	0.06377200
H	2.54796000	3.14529800	-0.64768600
H	-0.50399500	-0.74577200	0.22315000
H	-1.63045000	-0.63895100	1.54635800

IM1-5:

C	1.06373300	-1.88695900	1.46766200
C	0.70644300	-2.66929000	-0.91203200
C	1.81611400	-1.99677700	-1.23533800
C	2.56226400	-1.18329100	-0.21649400
H	0.15274200	-3.25459600	-1.63755100
H	1.80017100	-2.56572500	1.92659800
H	2.23542300	-2.02566400	-2.23527600
O	1.70900700	-0.78013000	0.84749900
H	0.42101800	-1.49350500	2.25327800
H	3.38458700	-1.78832400	0.20623600
O	3.05352100	-0.03922800	-0.84312900
H	3.53742100	0.47353800	-0.18182200
C	0.23495500	-2.69706300	0.48283900
O	-0.71565700	-3.36542000	0.85164500
C	-3.39382000	1.04339400	-0.75049900
C	-2.71405300	1.99990300	-0.10440700
C	-1.78049600	-0.00245100	0.49661600
N	-3.02726800	-0.23596400	-0.25117900
H	-4.20758400	1.14051700	-1.45311700
H	-2.83391200	3.07155800	-0.15679700
N	-1.85189900	1.42113200	0.85679700
C	-0.58197800	2.08394400	1.15861600
H	-0.81632800	3.04684600	1.62487600
H	-0.07194500	1.48130700	1.91624100
C	-2.94924200	-1.33329900	-1.20340800
H	-2.75833400	-2.26854400	-0.67307900
H	-3.89881300	-1.42605500	-1.73443200
H	-2.14340900	-1.18715700	-1.94240100
C	0.35336800	2.31304400	-0.03423200
H	-0.15776900	2.94449000	-0.77072400
H	0.57726300	1.36064800	-0.52129400
C	1.66784200	2.97222500	0.38545900
H	1.45465100	3.86720300	0.98323800
H	2.21427400	2.28581400	1.04227000
C	2.54699100	3.35661700	-0.80499500
H	2.04669700	4.09686500	-1.43798600
H	3.49865500	3.78739700	-0.47857800
H	2.76747300	2.48429400	-1.42630600
H	-0.90447200	-0.21047500	-0.15144900
H	-1.71133100	-0.63836900	1.38191200

HDPO:

C	0.70364600	-1.22822200	0.17919500
C	0.73818100	1.29020500	-0.07254000
C	-0.58363000	1.26865500	0.12922500
C	-1.32410500	-0.02538200	0.32129800
H	1.28825800	2.21452400	-0.21173700
H	0.72093900	-1.46003500	1.25660700
H	-1.17058800	2.17883400	0.19258200
O	-0.63175500	-1.10530200	-0.29626700
H	1.16905000	-2.05362500	-0.35850800
H	-1.40999800	-0.23672500	1.40275500
O	-2.57785500	0.08997400	-0.27122500
H	-3.08480400	-0.69670800	-0.03149600
C	1.51650100	0.04017000	-0.03419200
O	2.73255800	0.01297600	-0.10602200

Bmim*

C	-2.30991200	1.08876000	-0.39388900
C	-1.04092000	1.36608000	-0.00715700
C	-1.41070900	-0.85317900	0.42239000
N	-2.50898500	-0.25887100	-0.12494300
H	-3.07443600	1.71711600	-0.82008100
H	-0.47740200	2.28389500	-0.03381600
N	-0.51915600	0.17752400	0.48635900
C	0.85636000	0.03928700	0.96145000
H	1.08209000	0.88770000	1.61302800
H	0.89427600	-0.86807400	1.56479000
C	-3.76351700	-0.94791500	-0.39170900
H	-3.65185300	-1.98973100	-0.09870600
H	-4.57626000	-0.49688300	0.18190200
H	-4.00747600	-0.89614100	-1.45502100
C	1.86725400	-0.03580500	-0.18163400
H	1.78597400	0.86809100	-0.79571100
H	1.60807400	-0.88348100	-0.82586300
C	3.30250200	-0.18661500	0.32434100
H	3.54574800	0.65911700	0.97825600
H	3.37319800	-1.08776700	0.94469300
C	4.32244800	-0.26527600	-0.81123300
H	4.29495700	0.63850900	-1.42853900
H	5.34027200	-0.37501600	-0.42577300
H	4.11879100	-1.11992300	-1.46442100

BmimH

C	-2.19153400	-0.59459400	1.00232600
C	-1.02964000	-1.22561000	0.78651800
C	-1.30171000	0.27135900	-0.92087200
N	-2.54853300	0.16411500	-0.14597000
H	-2.88277500	-0.69681400	1.82489200
H	-0.53456200	-1.96754800	1.39467800
H	-1.48692900	0.27949700	-1.99729800
N	-0.54762400	-0.92084700	-0.50823800
C	0.89500300	-0.92061700	-0.74434500
H	1.25771000	-1.93237300	-0.53527900
H	1.05465000	-0.74488600	-1.81307700
C	-3.22921800	1.42988500	0.07102200
H	-3.55846300	1.84252200	-0.88588400
H	-4.11216900	1.26867200	0.69270500
H	-2.58124800	2.17456000	0.56438700
C	1.71274400	0.08684100	0.07404100
H	1.53321100	-0.09008800	1.14103200
H	1.37205900	1.10627700	-0.13628600
C	3.21049800	-0.01530600	-0.21896700
H	3.55003400	-1.03730500	-0.01146800
H	3.38154800	0.15414400	-1.28888800
C	4.04523800	0.97453700	0.59356600
H	3.91603000	0.80890600	1.66813100
H	5.11155100	0.87900100	0.36765600
H	3.75041400	2.00703900	0.37949700
H	-0.76624700	1.20654800	-0.64508200

Furfuryl alcohol:

C	-1.59862600	-0.62008400	0.13213800
H	-1.82290700	-1.02237000	1.12490000
C	-0.18133300	-0.15103900	0.06922100
C	0.38676700	1.08466600	0.10370600
O	0.78621900	-1.10805000	-0.04593800
C	1.80646500	0.87944100	0.00525600
H	-0.13647900	2.02306300	0.19309900
C	1.99302300	-0.46150200	-0.07951500
H	2.57728100	1.63461100	-0.00127900
H	2.86285800	-1.09146900	-0.16602200
H	-1.74348500	-1.43686300	-0.58761400
O	-2.51425800	0.44696600	-0.07707100
H	-2.35073800	0.79280700	-0.96384300

IM2-1:

C	0.13972500	-2.63818200	-0.35317300
O	-0.72401700	-2.63228400	0.75771700
C	-3.60684300	1.12917200	-0.56333600
C	-2.62602400	1.97923000	-0.16940100
C	-1.99226300	-0.11909100	0.45244400
N	-3.19677800	-0.13860200	-0.17739900
H	-4.54154300	1.31008600	-1.06752900
H	-2.53375000	3.04794000	-0.26798400
N	-1.65826800	1.19870100	0.44762200
C	-0.38976100	1.72478500	0.95231800
H	-0.59674800	2.66203000	1.47469400
H	-0.01071000	1.00794600	1.68066200
C	-3.96731000	-1.35366200	-0.40838000
H	-3.35536400	-2.20258100	-0.10986800
H	-4.88737600	-1.34232100	0.17973900
H	-4.21923100	-1.44255600	-1.46654200
C	0.63321300	1.94938800	-0.15946300
H	0.19786100	2.60611100	-0.92096500
H	0.84544600	0.99068300	-0.64443400
C	1.93011900	2.56308100	0.36882600
H	1.70310100	3.51317700	0.86673000
H	2.35375300	1.90402300	1.13435500
C	2.96174800	2.79657100	-0.73372400
H	2.57714900	3.48558600	-1.49277400
H	3.88460400	3.22471000	-0.33133800
H	3.21932500	1.85841300	-1.23425200
H	-0.32631300	-2.18846300	-1.24343200
H	-1.14280500	-1.72168300	0.77738800
H	0.35940200	-3.68144900	-0.61117200
C	1.42311200	-1.91803400	-0.07250900
C	1.98669000	-1.43037300	1.06584000
O	2.25063900	-1.68533900	-1.13667500
C	3.24951500	-0.85793800	0.68271400
H	1.55544200	-1.47222100	2.05245100
C	3.35945500	-1.03796200	-0.65749600
H	3.97039700	-0.37415700	1.32335000
H	4.11144000	-0.78691300	-1.38693400

TS2-1:

C	-0.57434300	-2.16891800	-0.04528200
O	-1.13533000	-2.51015200	1.05019900
C	-3.23244400	1.45597300	-0.68354600
C	-2.17114900	2.19575600	-0.28437800
C	-1.83351300	0.08174300	0.44423200
N	-3.07245900	0.17909600	-0.16124100
H	-4.09802800	1.72401800	-1.26523500
H	-1.93704500	3.23392100	-0.44660700
N	-1.34923800	1.37224800	0.47890000
C	-0.04277400	1.73895900	1.00913300
H	-0.14176300	2.70425500	1.51250700
H	0.22135800	0.99442000	1.76283800
C	-3.86611700	-0.98773600	-0.49381400
H	-3.66627600	-1.76364200	0.24518600
H	-4.92497000	-0.72880700	-0.48229900
H	-3.59873000	-1.37198600	-1.48383600
C	1.03847700	1.81156400	-0.06928600
H	0.74733900	2.56012700	-0.81468000
H	1.09560100	0.84976900	-0.58660300
C	2.40517500	2.16901900	0.51525700
H	2.32450000	3.10807000	1.07568800
H	2.69786200	1.39940600	1.23804700
C	3.48596500	2.30438100	-0.55625700
H	3.23874500	3.10122300	-1.26520600
H	4.45795300	2.54112000	-0.11356700
H	3.59441500	1.37489300	-1.12287800
H	-1.01960500	-0.81807600	-0.29585500
H	-1.70385000	-0.62182300	1.28944600
H	-0.93388500	-2.57552700	-1.01214100
C	0.88305700	-1.90634800	-0.05370000
C	1.80135400	-1.79342400	0.95175000
O	1.49875900	-1.70264200	-1.26262400
C	3.05895000	-1.50077400	0.32656400
H	1.59791500	-1.91328400	2.00344200
C	2.82053300	-1.45512400	-1.01049100
H	4.01149600	-1.34496700	0.80927200
H	3.44238500	-1.28059000	-1.87283500

IM2-2

C	0.04250000	-2.76018500	-0.57528800
O	-0.82263100	-3.27330000	0.11547500
C	-3.38685400	1.26555200	-0.26250500
C	-2.46010900	2.06029700	0.29105000
C	-1.68096900	-0.08026800	0.44828800
N	-3.08760100	-0.09372600	0.02450500
H	-4.31512300	1.53057000	-0.74514900
H	-2.44322700	3.13701700	0.36741800
N	-1.49888300	1.27725500	0.97151300
C	-0.12987400	1.77499600	1.09236800
H	-0.18440600	2.74594700	1.59511000
H	0.41111000	1.10022500	1.76325900
C	-3.39073900	-1.08531400	-0.99447300
H	-3.21150500	-2.08699700	-0.59891300
H	-4.44131300	-1.00742600	-1.28115300
H	-2.76669300	-0.95818800	-1.89596900
C	0.64747400	1.92759700	-0.22076300
H	0.05916500	2.53914200	-0.91522500
H	0.77587300	0.95038800	-0.69616100
C	2.01972200	2.56841500	-0.00862100
H	1.89001300	3.56066200	0.44001400
H	2.58765000	1.97381200	0.71539400
C	2.81975400	2.69338400	-1.30479100
H	2.29283000	3.31821600	-2.03340000
H	3.80167700	3.14286800	-1.12860300
H	2.97899700	1.71287100	-1.76429400
H	-1.02528300	-0.26647200	-0.43131700
H	-1.46366100	-0.84570300	1.19641900
H	0.00733000	-2.80015000	-1.67982400
C	1.18594700	-2.05640200	-0.03706000
C	1.59386200	-1.76418700	1.24023200
O	2.08793600	-1.53722600	-0.93737600
C	2.80288900	-1.02794900	1.12025600
H	1.08018000	-2.04481500	2.14582100
C	3.05244500	-0.91961700	-0.21914200
H	3.40910000	-0.62554800	1.91604400
H	3.84395300	-0.45482200	-0.78369400

Furfural:

C	-0.25798200	0.26544700	-0.00012000
C	0.77108200	1.17381900	-0.00013600
O	0.25131700	-1.00806400	-0.00003400
C	1.97701800	0.42063800	0.00023900
H	0.66344400	2.24758400	-0.00019700
C	1.59867500	-0.89165300	-0.00007200
H	2.98845700	0.79463200	0.00047400
H	2.15537300	-1.81527700	-0.00006500
C	-1.69065700	0.46762900	-0.00023100
H	-1.96882900	1.53916400	0.00021000
O	-2.52972400	-0.41460900	0.00022200

IM3-1:

C	0.29174300	0.48110100	-0.04022200
C	-0.69769900	0.49441000	1.08588900
O	-0.39321400	-0.24309400	-1.10086700
C	-1.79399400	-0.25711200	0.69322200
H	-0.50619900	0.97722200	2.03170100
C	-1.57554900	-0.68315600	-0.60495900
H	-2.68037300	-0.47069000	1.27210800
H	-2.18966500	-1.26667100	-1.27341700
C	1.52259200	-0.32624800	0.39912800
H	2.19084400	0.25137300	1.06668500
O	1.71851800	-1.47855600	0.11273800
O	0.76028100	1.71178300	-0.48972600
H	0.01815000	2.33373200	-0.47257700

TS3-1

C	-0.07812200	-0.81879100	0.00443200
C	0.58236100	-0.39041600	1.15100400
O	0.54201000	-0.28999400	-1.10314600
C	1.54986400	0.55643400	0.71541700
H	0.34298700	-0.68932400	2.15910600
C	1.47059400	0.59788400	-0.64481800
H	2.20517800	1.15172500	1.33176800
H	1.97918000	1.18046800	-1.39498900
C	-1.60259800	0.69103700	0.23071400
H	-2.10790600	0.47071900	1.19998500
O	-1.36807800	1.77597200	-0.19827600
O	-0.86614000	-1.89234300	-0.21817400
H	-0.41436700	-2.67956100	0.12039900

IM3-2:

C	-0.67517300	0.06606100	0.00008900
C	0.12802100	1.16641500	-0.00037800
O	0.04420500	-1.07422300	0.00017300
C	1.47277900	0.64877100	0.00024000
H	-0.18955700	2.19680300	-0.00064500
C	1.37592000	-0.70182700	-0.00041300
H	2.38657000	1.22294200	0.00044900
H	2.08482200	-1.51181900	-0.00068000
O	-2.00026000	-0.13903400	0.00025100
H	-2.44268000	0.72160400	0.00025300

IM3-3:

C	-0.83759700	0.02573300	-0.00018400
C	0.05515100	1.20204000	-0.00005400
O	-0.05392300	-1.09795000	0.00007300
C	1.31868000	0.77757700	-0.00002600
H	-0.31906000	2.21471600	-0.00004400
C	1.33418100	-0.71424700	0.00009100
H	2.22150400	1.37293200	0.00000600
H	1.81817800	-1.13064000	-0.88818700
O	-2.04123100	-0.03619200	0.00002600
H	1.81811900	-1.13049400	0.88847100

IM3-4:

C	-0.50153500	1.01565900	-0.00006000
C	0.82991500	0.87827100	0.00015600
C	1.61498600	-0.39342700	0.00012700
H	-0.94755700	2.00235000	-0.00000400
H	1.43746700	1.77943300	0.00037800
H	1.35107300	-0.99461300	-0.87957900
O	2.99964600	-0.04231800	-0.00009300
H	3.50240200	-0.86448400	-0.00047800
O	-2.71418800	0.26781900	-0.00004400
H	-3.25980100	-0.53623900	0.00024500
H	1.35133200	-0.99445400	0.88003300
C	-1.42157700	-0.12942900	-0.00043100
O	-1.10616400	-1.30280600	0.00021800

IM3-5:

C	3.92646400	1.12671500	0.27154900
C	2.90404900	1.86323700	-0.23003600
C	2.23598400	-0.31583200	-0.23849900
N	3.49439500	-0.19181900	0.25996100
H	4.90272900	1.41587500	0.62343000
H	2.80935300	2.92360800	-0.39460800
N	1.89121500	0.96515500	-0.53694200
C	0.56872600	1.36580900	-1.01789700
H	0.70308200	2.17110000	-1.74398700
H	0.13818300	0.50931300	-1.53710600
C	4.30159100	-1.31804800	0.71022800
H	3.66241300	-2.19745000	0.75734700
H	5.12511400	-1.50340900	0.01719500
H	4.70750400	-1.11326600	1.70219900
C	-0.34530700	1.81718600	0.11982700
H	0.15277100	2.61327100	0.68455100
H	-0.49084700	0.97821000	0.80796300
C	-1.70117100	2.31436800	-0.38179000
H	-1.54806900	3.15407800	-1.06972800
H	-2.18442700	1.52372300	-0.96460300
C	-2.62171600	2.74792100	0.75886400
H	-2.19305000	3.59219300	1.30856600
H	-3.60244900	3.05617000	0.38576700
H	-2.77259100	1.93346400	1.47334700
H	-0.45533000	-1.61193200	0.96927900
H	1.33491600	-1.94010000	-0.27821400
C	-1.35602900	-2.10704100	-0.91383200
H	-0.99114800	-2.49987600	-1.86005200
C	-2.50823600	-1.42237300	-0.93128000
H	-3.03337200	-1.27078500	-1.86663300
C	-3.13088500	-0.83251600	0.26051800
O	-2.73688100	-0.91349300	1.40714400
C	-0.46728800	-2.43951900	0.25239300
H	-0.90624800	-3.29520600	0.78316000
O	0.83253800	-2.79716600	-0.16853400
O	-4.25725900	-0.15013000	-0.06117300
H	-4.60208100	0.23454500	0.76123400

TS3-2:

C	-2.97403500	1.05155600	-1.34052600
C	-2.11515400	2.00594900	-0.90696100
C	-1.90498000	0.22964300	0.47055800
N	-2.90164000	-0.00894500	-0.45088700
H	-3.64612400	1.03970000	-2.18177800
H	-1.89227100	2.98178000	-1.30325400
N	-1.51367800	1.53435900	0.25397800
C	-0.32594000	2.10812600	0.87755100
H	-0.51778800	3.16334500	1.08455200
H	-0.18972100	1.60061100	1.83430600
C	-3.54765100	-1.30112200	-0.59347800
H	-3.42100000	-1.84682300	0.34144200
H	-4.60763300	-1.16525000	-0.81105500
H	-3.08348300	-1.87681800	-1.39982200
C	0.91695500	1.94144600	-0.00110000
H	0.81050400	2.56528200	-0.89512700
H	0.96767300	0.90319100	-0.33948000
C	2.20584000	2.30116700	0.73621400
H	2.14194800	3.33107800	1.10675100
H	2.30191500	1.65821800	1.61861900
C	3.44378100	2.14661400	-0.14683500
H	3.39311900	2.81145500	-1.01525400
H	4.35852600	2.38612800	0.40323200
H	3.53436100	1.12163100	-0.51861000
H	-0.82337100	-0.69168600	0.29380100
H	-2.00255700	-0.22399900	1.47642100
C	0.89648700	-1.51522200	1.36554500
H	0.96885000	-1.24803400	2.41825000
C	2.02923100	-1.45289600	0.63383900
H	2.95830100	-1.16054700	1.10765600
C	2.09419800	-1.71016600	-0.80206000
O	1.16334500	-1.94534100	-1.55014700
C	-0.47999300	-1.91244600	0.97684700
H	-0.53530600	-2.52560400	0.06279700
O	-1.35602100	-2.05127400	1.90350800
O	3.37140300	-1.63677400	-1.26574300
H	3.32902600	-1.79034000	-2.22359700

IM3-6:

C	3.25102400	0.77467700	1.14829600
C	2.64869000	1.80330000	0.53449200
C	1.95070400	-0.10418100	-0.51778100
N	3.04796100	-0.41601000	0.40436700
H	3.91424500	0.77970100	1.99970900
H	2.69632500	2.85658500	0.76532300
N	2.00808400	1.35398500	-0.64279200
C	0.78805400	2.02014200	-1.09679600
H	1.05151400	3.04439600	-1.37949100
H	0.46419200	1.51446200	-2.01188200
C	2.88576300	-1.66362400	1.13249400
H	2.86642600	-2.50020800	0.43052100
H	3.72805600	-1.80360900	1.81255000
H	1.95051700	-1.68314600	1.71679200
C	-0.36542700	2.05371800	-0.08599600
H	-0.05803900	2.63941500	0.78792300
H	-0.57491200	1.04186000	0.27383300
C	-1.63741800	2.65253800	-0.68677300
H	-1.41138400	3.64415300	-1.09739900
H	-1.95904300	2.03473700	-1.53357600
C	-2.77779900	2.76825100	0.32415300
H	-2.49467700	3.41187900	1.16341500
H	-3.67599400	3.19383400	-0.13357400
H	-3.04496700	1.78938500	0.73206800
H	0.98177000	-0.42988200	-0.07354300
H	2.05992200	-0.60937700	-1.48064800
C	-1.24381500	-1.63144700	-1.37127100
H	-1.18521600	-1.44348100	-2.43926700
C	-2.24528300	-1.07428800	-0.67372000
H	-2.98027300	-0.46911700	-1.19072500
C	-2.45008600	-1.20550400	0.78774000
O	-1.65190800	-1.61182200	1.60326900
C	-0.19934100	-2.53975100	-0.85479400
H	-0.25745700	-2.82299200	0.20432800
O	0.67235900	-2.97695200	-1.58554900
O	-3.68618200	-0.78630300	1.13062100
H	-3.76788200	-0.87680200	2.09562400

IM3-7:

C	-0.96690500	0.90270700	-0.00020100
H	-1.54790300	1.82044600	-0.00031900
C	0.37085800	0.99078100	-0.00001600
H	0.83309700	1.97100100	-0.00001100
C	1.32006000	-0.15159000	0.00019600
O	1.04739100	-1.33096900	-0.00021200
C	-1.78130000	-0.33134200	-0.00028300
H	-1.24405400	-1.28939000	0.00057900
O	-2.99596900	-0.28013500	0.00027100
O	2.58950500	0.30000700	0.00013800
H	3.17515700	-0.47662100	0.00000200

HFO:

C	0.68888800	1.25146700	-0.12132300
O	0.10739200	-0.94608200	0.28263100
C	-0.61568300	1.26536500	0.13252700
H	1.34247000	2.07368300	-0.37236800
C	-1.09141100	-0.13449400	0.41907700
H	-1.29283700	2.10844800	0.14029500
O	-2.10176600	-0.59288200	-0.41184000
H	-1.86513800	-0.37607300	-1.32565800
C	1.17367300	-0.14577900	-0.02123200
O	2.28553200	-0.58197400	-0.16330000
H	-1.44656100	-0.25790600	1.44351600

meso-PmPD/GO-furfuryl alcohol-BmimH

C	2.19044900	-3.29608800	-0.58696800
C	1.59562000	-2.03013100	-0.50734500
C	2.37323000	-0.90242300	-0.26060200
C	3.77075800	-1.03802500	-0.10813600
C	4.35489100	-2.29590200	-0.20898700
C	3.56481500	-3.43021500	-0.43838600
C	3.99825900	1.38669000	-0.08223200
C	2.59892600	1.51550500	-0.22912900
C	2.04601300	2.77515000	-0.44715800
H	0.97174800	2.85775600	-0.56560800
C	2.86473500	3.91054800	-0.50240500
C	4.24016400	3.78243600	-0.36223600
C	4.80545300	2.51583100	-0.16126000
H	1.56981900	-4.16541100	-0.76978100
H	0.52556300	-1.91219600	-0.64433100
H	5.43000700	-2.38633800	-0.09377600
H	4.03474000	-4.40471500	-0.50224700
H	2.41593300	4.88439100	-0.66024600
H	4.88319100	4.65351700	-0.40812900
H	5.87884200	2.40189200	-0.05126100
N	4.51693100	0.11228100	0.17818200
H	5.52261400	0.01804600	0.13797300
N	1.81459000	0.36808200	-0.12560800
H	0.80855400	0.47601500	-0.25748900
C	-0.91928700	1.36134600	1.46413300
H	0.01815600	1.87004200	1.69183600
C	-0.98252200	0.08737200	2.22933000
C	-0.04409400	-0.81922000	2.62101200
O	-2.23103800	-0.37013400	2.57709800
C	-0.74826100	-1.90151600	3.24626900
H	1.01966200	-0.72727900	2.47093800
C	-2.06724800	-1.57804300	3.19206800
H	-0.32546700	-2.79459600	3.67953000
H	-2.96814100	-2.06205500	3.53151400
H	-1.74090200	2.01506000	1.77447300
O	-0.94028600	1.16665100	0.04406100
H	-1.73353200	0.62037500	-0.19510500
C	-2.80845700	-0.31925400	-2.23010000
C	-2.00153000	-1.31386300	-2.61399800
C	-2.71346800	-1.83064000	-0.51752300
N	-3.06619400	-0.42644400	-0.82017600
H	-3.13807100	0.54298000	-2.78497100

H	-1.53382100	-1.47562700	-3.57346400
H	-2.34160700	-1.94096100	0.49977600
N	-1.68975300	-2.14793100	-1.51806400
C	-4.41075700	-0.03851600	-0.34456800
H	-4.65168800	-0.68569700	0.50136000
H	-5.15104100	-0.24454600	-1.12897900
H	-3.60022400	-2.48343400	-0.65932100
C	-1.49291800	-3.56745900	-1.76672600
H	-2.42115900	-4.06539300	-2.08922400
H	-0.73493000	-3.69875300	-2.54039900
H	-1.13902400	-4.05178400	-0.85431600
C	-4.50636900	1.41365900	0.11884300
H	-5.53508200	1.57732800	0.46200600
H	-3.86370700	1.54661900	0.99487900
C	-4.15515500	2.46812400	-0.93282300
H	-3.11427400	2.34031800	-1.24281900
H	-4.77285400	2.31353400	-1.82552600
C	-4.34917600	3.89246400	-0.41168000
H	-3.72102700	4.07690200	0.46597800
H	-4.08583400	4.63456500	-1.17104000
H	-5.38926800	4.06840200	-0.11821000

meso-PA/PmPD/GO-furfuryl alcohol-BmimH

C	-2.24433100	3.22713700	1.12332000
C	-1.65416100	1.99821600	1.43717100
C	-0.27265100	1.84328200	1.38582700
C	0.53581900	2.94864500	1.04613900
C	-0.05575500	4.16987700	0.74325200
C	-1.44884100	4.31017500	0.77183600
C	2.50857100	1.68712400	1.68636700
C	1.69603200	0.57579500	1.99727500
C	2.26387900	-0.53360600	2.61912400
H	1.63276200	-1.38130200	2.85680000
C	3.62883000	-0.55209800	2.92856900
C	4.42418500	0.54867400	2.63453000
C	3.85755800	1.67251600	2.02170700
H	-3.32274800	3.32390800	1.15646600
H	-2.26607100	1.15101700	1.71928400
H	0.57881000	5.00992500	0.48103600
H	-1.89612700	5.26508900	0.52260100
H	4.05634500	-1.42926600	3.39992400
H	5.48097900	0.54488700	2.87386000

H	4.46727700	2.53699400	1.78074800
N	1.92350300	2.75408900	0.98873800
H	2.48561200	3.59077000	0.90580800
N	0.34806500	0.61447800	1.62862000
H	-0.24007700	-0.14377200	1.98036700
H	2.29736100	-1.47905400	-1.60135600
P	0.39314600	-3.09732700	-0.55973800
O	1.77777100	-2.85089900	-1.10758300
O	-0.52046400	-1.82041600	-0.98523100
H	-1.27822000	-1.66868500	-0.39459000
O	-0.21097200	-4.32130300	-1.44679100
O	0.19701500	-3.38434600	0.91171300
C	-2.32920600	-2.15889700	2.65687200
H	-2.42692000	-3.21885400	2.39412100
C	-3.19446500	-1.33717800	1.76470700
C	-4.31650000	-0.59492900	1.95740500
O	-2.84878800	-1.28666900	0.43129900
C	-4.68973600	-0.05252300	0.67885700
H	-4.81709200	-0.44471600	2.90119700
C	-3.76585900	-0.49027800	-0.21023800
H	-5.52741900	0.59104100	0.46088300
H	-3.60597700	-0.34475200	-1.26489500
H	-2.69684700	-2.03493400	3.67840700
O	-0.95771300	-1.77785500	2.60098700
H	-0.49287600	-2.40083000	1.97919200
C	2.97669500	-0.39369700	-3.26439500
C	1.96348800	0.27902700	-3.81988000
C	1.60919100	0.50019900	-1.57664400
N	2.70043000	-0.50979600	-1.81924600
H	3.79582800	-0.94549700	-3.69051800
H	1.81989700	0.49120000	-4.86946300
H	0.91287000	0.11138800	-0.84046100
N	1.01020900	0.67034500	-2.89968200
C	0.17509600	1.84981900	-3.11184700
H	0.66897600	2.73968800	-2.69305900
H	0.09323700	1.99778700	-4.19080000
C	3.87904100	-0.34568100	-0.92954800
H	3.55377700	-0.47744700	0.09923500
H	4.60627500	-1.11413200	-1.18714900
H	4.30455200	0.64508300	-1.07508700
C	-1.21884400	1.68615500	-2.50981200
H	-1.14195200	1.51280200	-1.43197600
H	-1.67777500	0.79102400	-2.94393200
C	-2.10679900	2.90469700	-2.77012000

H	-1.68072900	3.77648500	-2.26156800
H	-2.09741000	3.13485600	-3.84251000
C	-3.54938600	2.69618700	-2.30918600
H	-3.59159000	2.46361100	-1.24268600
H	-4.15438500	3.59146100	-2.48138900
H	-4.01762200	1.86794900	-2.85080700
H	2.06510200	1.43307800	-1.22606900
C	-1.57212000	-4.72145400	-1.22961900
H	-1.76206100	-5.57063500	-1.88530500
H	-1.73099900	-5.02125400	-0.19049300
H	-2.26057000	-3.91003100	-1.48390800

6. References

- (1) Marcano, D. C.; Kosynkin, D. V.; Berlin, J. M.; Sinitskii, A.; Sun, Z.; Slesarev, A.; Alemany, L.B.; Lu, W.; Tour, J. M. Improved Synthesis of Graphene Oxide. *ACS Nano* **2010**, *4*, 4806-4814, DOI 10.1021/nn1006368.
- (2) Kang, X.; Wang, B.; Hu, K.; Lyu, K.; Han, X.; Spencer, B. F.; Frogley, M. D.; Tuna, F.; McInnes, E. J. L.; Dryfe, R. A. W.; Han, B.; Yang, S.; Schröder, M. Quantitative electro-reduction of CO₂ to liquid fuel over electrosynthesized metal-organic frameworks. *J. Am. Chem. Soc.* **2020**, *142*, 17384-17392, DOI 10.1021/jacs.0c05913.
- (3) Snook, G. A.; Best, A. S.; Pandolfo, A. G.; Hollenkamp, A. F. Evaluation of a Ag/Ag⁺ reference electrode for use in room temperature ionic liquids. *Electrochem. Commun.* **2006**, *8*, 1405-1411, DOI 10.1016/j.elecom.2006.07.004.
- (4) Frisch, M. J.; Trucks, G. W.; Schlegel, H. B.; Scuseria, G. E.; Robb, M. A.; Cheeseman, J. R.; Scalmani, G.; Barone, V.; Mennucci, B.; Petersson, G. A.; Nakatsuji, H.; Caricato, M.; Li, X.; Hratchian, H. P.; Izmaylov, A. F.; Bloino, J.; Zheng, G.; Sonnenberg, J. L.; Hada, M.; Ehara, M.; Toyota, K.; Fukuda, R.; Hasegawa, J.; Ishida, M.; Nakajima, T.; Honda, Y.; Kitao, O.; Nakai, H.; Vreven, T.; Montgomery, J. A., Jr.; Peralta, J. E.; Ogliaro, F.; Bearpark, M.; Heyd, J. J.; Brothers, E.; Kudin, K. N.; Staroverov, V. N.; Kobayashi, R.; Normand, J.; Raghavachari, K.; Rendell, A.; Burant, J. C.; Iyengar, S. S.; Tomasi, J.; Cossi, M.; Rega, N.; Millam, N. J.; Klene, M.; Knox, J. E.; Cross, J. B.; Bakken, V.; Adamo, C.; Jaramillo, J.; Gomperts, R.; Stratmann, R. E.; Yazyev, O.; Austin, A. J.; Cammi, R.; Pomelli, C.; Ochterski, J. W.; Martin, R. L.; Morokuma, K.; Zakrzewski, V. G.; Voth, G. A.; Salvador, P.; Dannenberg, J. J.; Dapprich, S.; Daniels, A. D.; Farkas, Ö.; Foresman, J. B.; Ortiz, J. V.; Cioslowski, J.; Fox, D. J. Gaussian 16, Revision A.03. Gaussian Inc., Wallingford, CT, **2016**.
- (5) Lee, C. T.; Yang, W. T.; Parr, R. G. Development of the Colle-Salvetti correlation-energy formula into a functional of the electron density. *Phys. Rev. B: Condens. Matter Mater. Phys.* **1988**, *37*, 785-789, DOI 10.1103/PhysRevB.37.785.

- (6) Miehlisch, B.; Savin, A.; Stoll, H.; Preuss, H. Results obtained with the correlation energy density functionals of Becke and Lee, Yang and Parr. *Chem. Phys. Lett.* **1989**, *157*, 200-206, DOI 10.1016/0009-2614(89)87234-3.
- (7) Stephens, P. J.; Devlin, F. J.; Chabalowski, C. F.; Frisch, M. J. Ab Initio Calculation of Vibrational Absorption and Circular Dichroism Spectra Using Density Functional Force Fields. *J. Phys. Chem.* **1994**, *98*, 11623-11627, DOI 10.1021/j100096a001.
- (8) Marenich, A. V.; Cramer, C. J.; Truhlar, D. G. Universal Solvation Model Based on Solute Electron Density and on a Continuum Model of the Solvent Defined by the Bulk Dielectric Constant and Atomic Surface Tensions. *J. Phys. Chem. B.* **2009**, *113*, 6378-6396, DOI 10.1021/jp810292n.
- (9) Zhang L.; Wang T.; Wang H.; Meng, Y.; Yu, W.; Chai, L. Graphene@poly(*m*-phenylenediamine) hydrogel fabricated by a facile post-synthesis assembly strategy. *Chem. Commun.* **2013**, *49*, 9974-9976, DOI 10.1039/c3cc45261a.
- (10) Liu, Z.; Shang, S.; Chiu, K.; Jiang, S.; Dai, F. Fabrication of conductive and flame-retardant bifunctional cotton fabric by polymerizing pyrrole and doping phytic acid. *Polym. Degrad. Stabil.* **2019**, *167*, 277-282, DOI 10.1016/j.polymdegradstab.2019.06.029.
- (11) Wu, H.; Song, J.; Xie, C.; Hu, Y.; Ma, J.; Qian, Q.; Han, B. Design of naturally derived lead phytate as an electrocatalyst for highly efficient CO₂ reduction to formic acid. *Green Chem.* **2018**, *20*, 4602-4606, DOI 10.1039/c8gc02457j.
- (12) Yang, W.; Liu, H.; Li, Y.; He, D. Interaction mechanism of Ni(NO₃)₂ · 6H₂O and P123 in preparing highly-dispersed Ni/SBA-15 catalytic materials. *Micropor. Mesopor. Mat.* **2016**, *228*, 174-181, DOI 10.1016/j.micromeso.2016.03.044.
- (13) Zhang, K.; Nelson, A. M.; Talley, S. J.; Chen, M.; Margareta, E.; Hudson, A. G.; Moore, R. B.; Long, T. E. Non-isocyanate poly(amide-hydroxyurethane)s from sustainable resources. *Green Chem.* **2016**, *18*, 4667-4681, DOI 10.1039/c6gc01096b.
- (14) Mai, Y.; Zhou, Y.; Yan, D. Synthesis and size-controllable self-assembly of a novel amphiphilic hyperbranched multiarm copolyether. *Macromolecules* **2005**, *38*, 8679-8686, DOI 10.1021/ma051377y.

(15) Burattini, S.; Greenland, B. W.; Merino, D. H.; Weng, W.; Seppala, J.; Colquhoun, H. M.; Hayes, W.; Mackay, M. E.; Hamley, I. W.; Rowan, S. J. A healable supramolecular polymer blend based on aromatic π - π stacking and hydrogen-bonding interactions. *J. Am. Chem. Soc.* **2010**, *132*, 12051-12058, DOI 10.1021/ja104446r.