

Supplementary Information (SI) for

**Improved Conductivity of a New Co(II)-MOF by Assembled Acetylene
Black for Efficient Hydrogen Evolution Reaction**

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Table S1. Summary of crystallographic data for CTGU-9.

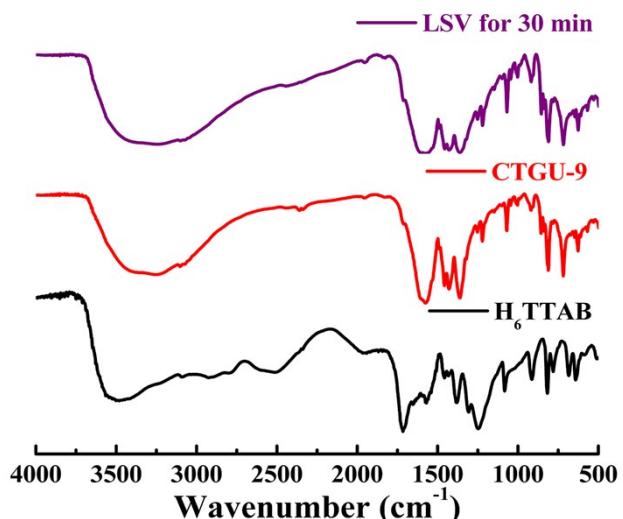
Compound	CTGU-9
Empirical formula	C ₁₉ H ₁₂ N ₃ O ₇ Co _{1.5}
Formula weight	482.71
Temperature	293(2) K
Crystal system	Monoclinic
Space group	P12/n1
a (Å)	9.3083(2)
b (Å)	10.4819(2)
c (Å)	22.1487(4)
α (°)	90
β (°)	96.098(2)
γ (°)	90
Z	4
V (Å ³)	2148.8(1)
ρ _{calc} (mg m ⁻³)	1.49202
μ (mm ⁻¹)	9.580
F(000)	974.0
θ _{min-max} (°)	9.344 to 133.186
Reflections collected	10644
Independent	3795
GOF on F ²	1.057
R(int)	0.067
R ₁ , wR ₂ (I > 2σ(I)) ^a	0.0612, 0.1620
R ₁ , wR ₂ (all data) ^b	0.0670, 0.1668
CCDC No.	1583330

^a R₁ = Σ(|F_o| - |F_c|)/Σ|F_o| ; ^bwR₂ = {Σ[w(|F_o|² - |F_c|²)²]/Σ[w(|F_o|²)²]}

Table S2. Selected bond lengths [\AA] and angles [°] for **CTGU-9**.

Co(1)-O(2)#1	2.048(2)	Co(1)-O(2)	2.048(2)
Co(1)-O(1)	2.139(2)	Co(1)-O(1)#1	2.139(2)
Co(1)-N(1)#1	2.225(3)	Co(1)-N(1)	2.225(3)
Co(2)-O(6)#2	1.948(2)	Co(2)-O(3)	1.946(3)
Co(2)-O(4)#3	1.967(3)	Co(2)-N(2)	2.032(3)
O(6)-Co(2)#4	1.948(2)	O(4)-Co(2)#3	1.967(3)
O(2)#1-Co(1)-O(2)	180.0	O(2)-Co(1)-O(1)	84.55(9)
O(2)-Co(1)-O(1)	95.45(9)	O(2)-Co(1)-O(1)#1	95.45(9)
O(2)#1-Co(1)-N(1)#1	88.83(11)	O(2)#1-Co(1)-N(1)	91.17(11)
O(1)#1-Co(1)-O(1)	180.0	O(1)#1-Co(1)-N(1)	96.07(10)
O(1)-Co(1)-N(1)	83.93(14)	N(1)#1-Co(1)-N(1)	180.0
O(6)#2-Co(2)-O(4)#3	100.17(10)	O6(2)-Co(2)-N2	103.38(12)
O3-Co(2)-O(6)#2	124.40(12)	O(3)-Co(2)-O(4)#3	95.19(11)
O(3)-Co(2)-N(2)	112.84(12)	O(4)#3-Co(2)-N2	112.64(12)
C(8)-O(6)-Co(2)#4	125.3(2)	C(1)-O(3)-Co(2)	124.5(2)
C(9)-O(4)-Co(2)#3	134.3(2)	C(1)-O(1)-Co(1)	138.1(2)
C(15)-N(2)-Co(2)	120.1(2)	C(19)-N(2)-Co(2)	122.4(3)
C(10)-N(1)-Co(1)	122.3(3)	C(14)-N(1)-Co(1)	122.3(3)
C(14)-N(1)-Co(1)	122.3(11)	C(14)-N(1)-Co(1)	122.3(3)

Symmetry codes: #1:1-x,1-y,1-z; #2:-1+x,+y,+z; #3:1-x,2-y,1-z; #4:1+x,+y,+z; #5:3/2-x,+y,3/2-z; #6: 1/2-x,+y,1/2-z.

**Figure S1.** Infrared spectra for H₆TTAB and **CTGU-9** before and after HER tests.

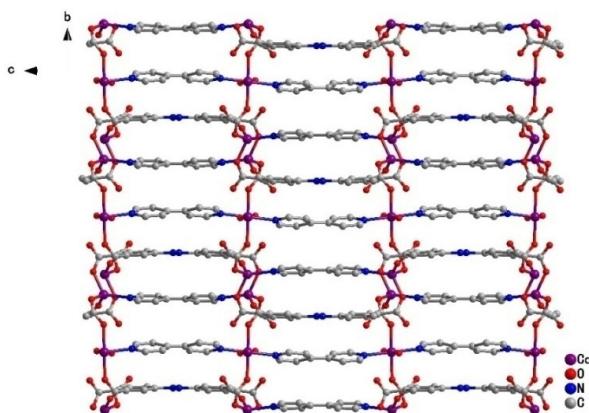


Figure S2. View of the novel 3D frameworks of CTGU-9 along the a axis.

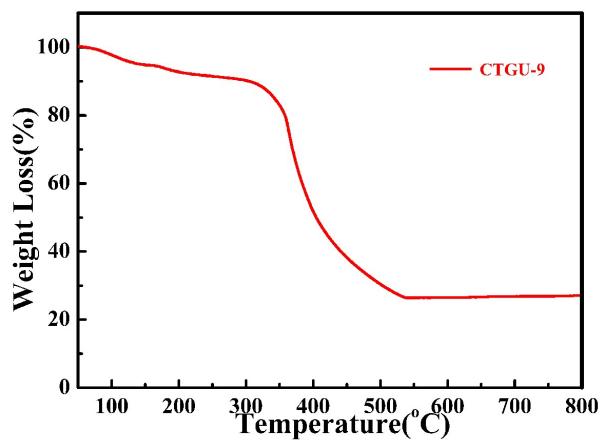


Figure S3. Thermal gravimetric analysis of CTGU-9.

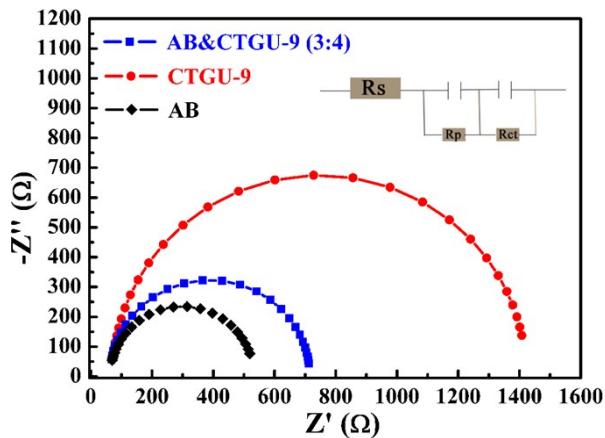


Figure S4. Nyquist plots of different catalysts over the frequency ranging from 1000 kHz to 0.1 Hz at a potential of -0.128V vs. RHE. (Inset: equivalent circuit used to fit the Nyquist plots).

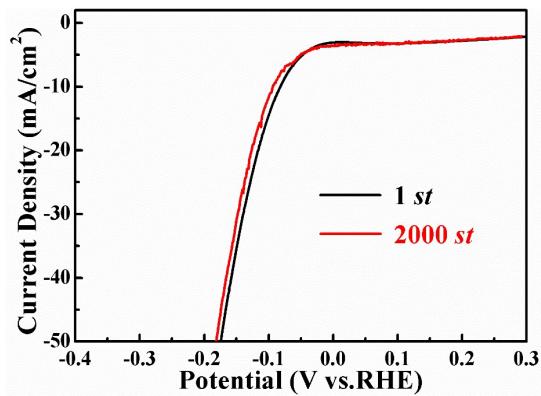


Figure S5. LSV curves of **AB&CTGU-9 (3:4)** before and after 2000 CV cycles.

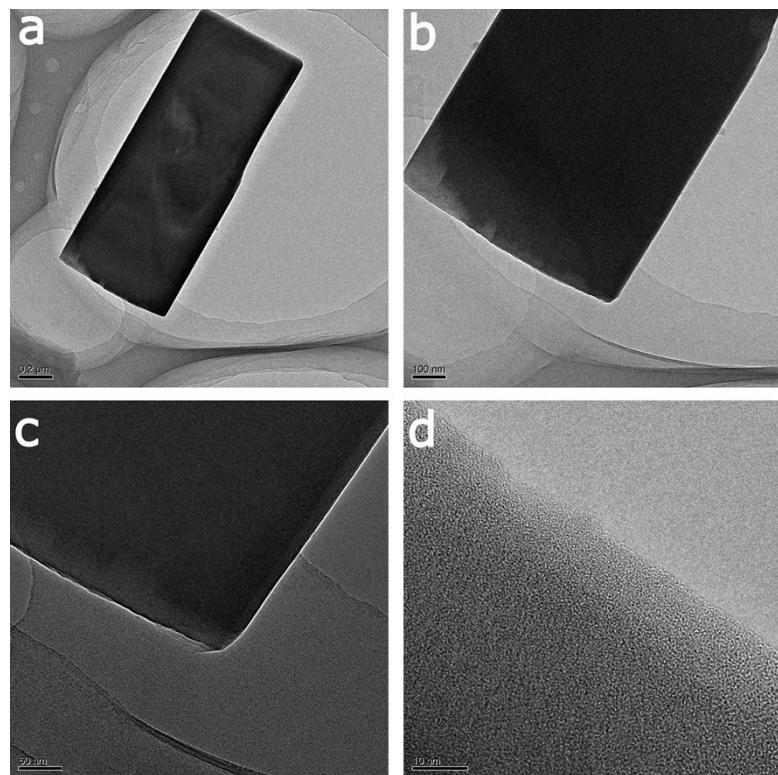


Figure S6. (a-d) TEM images of **CTGU-9**.

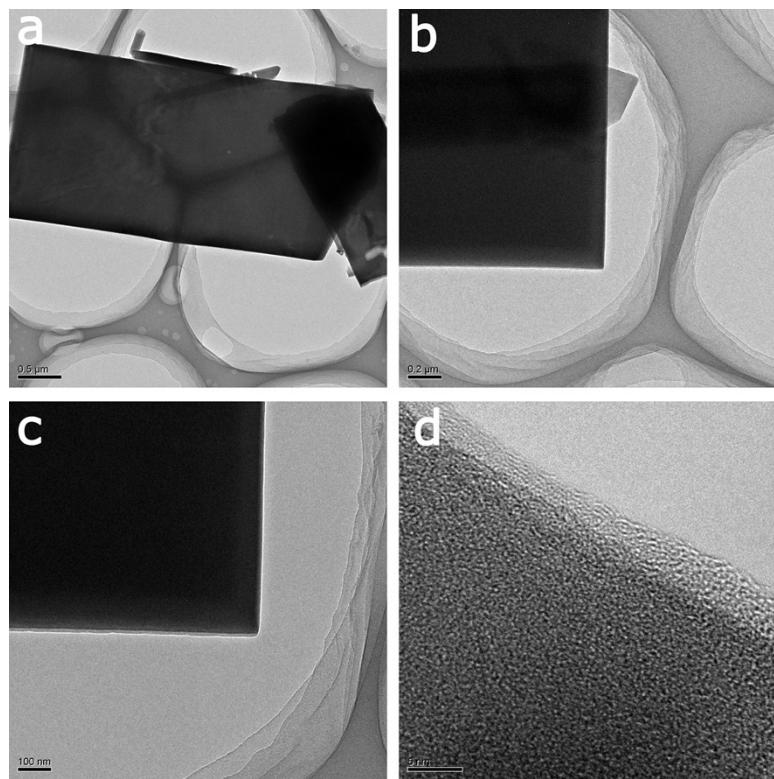


Figure S7. (a-d) TEM images of CTGU-9 after catalyzing water for 30 min.

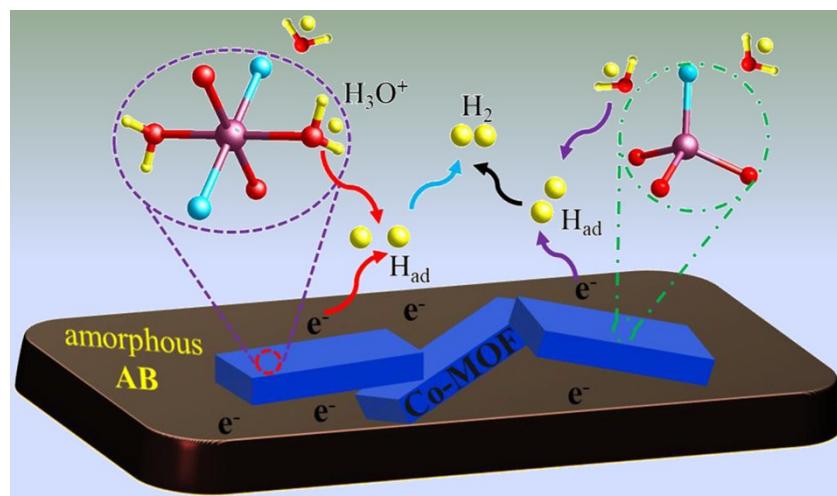


Figure. S8 The possible mechanism for AB&CTGU-9 catalyst.