

Supporting Information

# Seeded Growth of Gold-based Nanoscale Homojunctions *via* Controlled Etching-Regrowth and Their Applications for Methanol Oxidation Reaction

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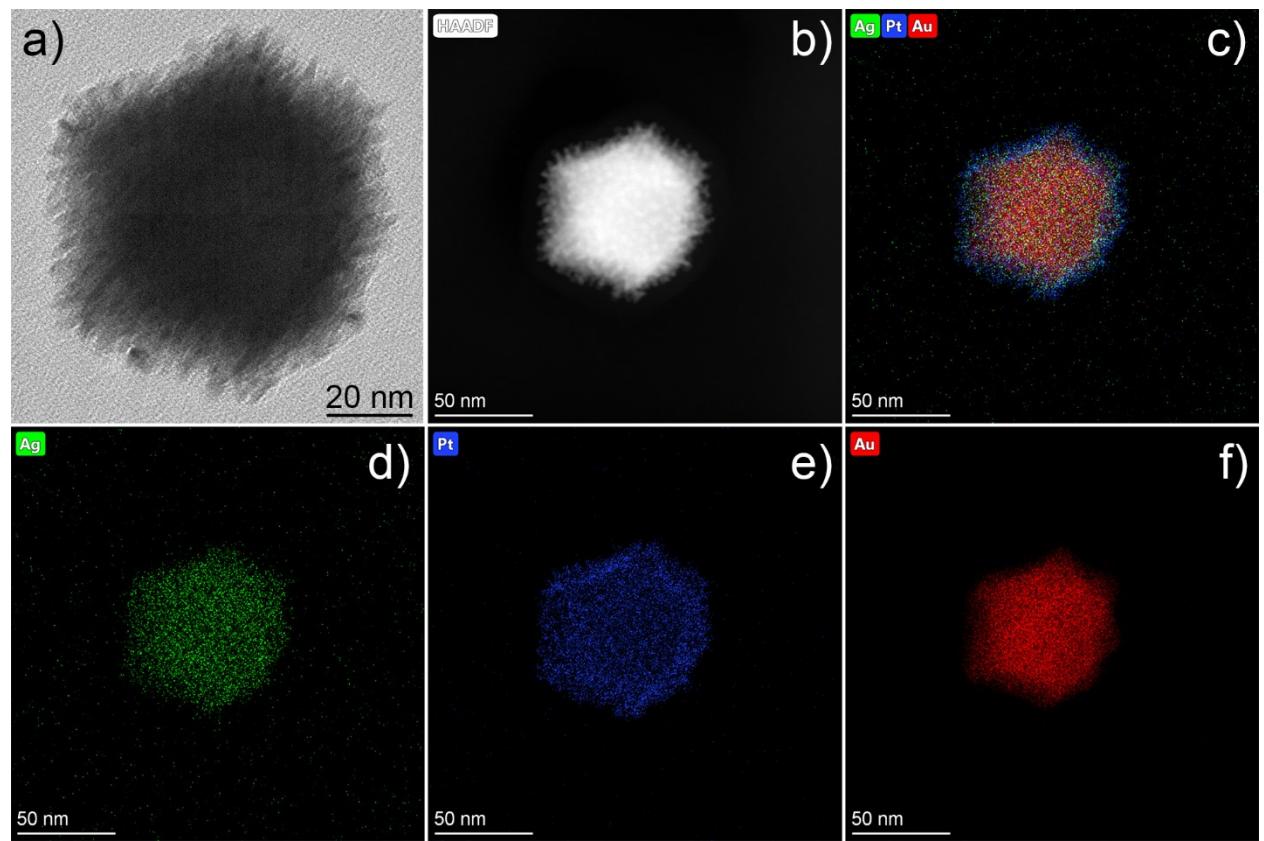
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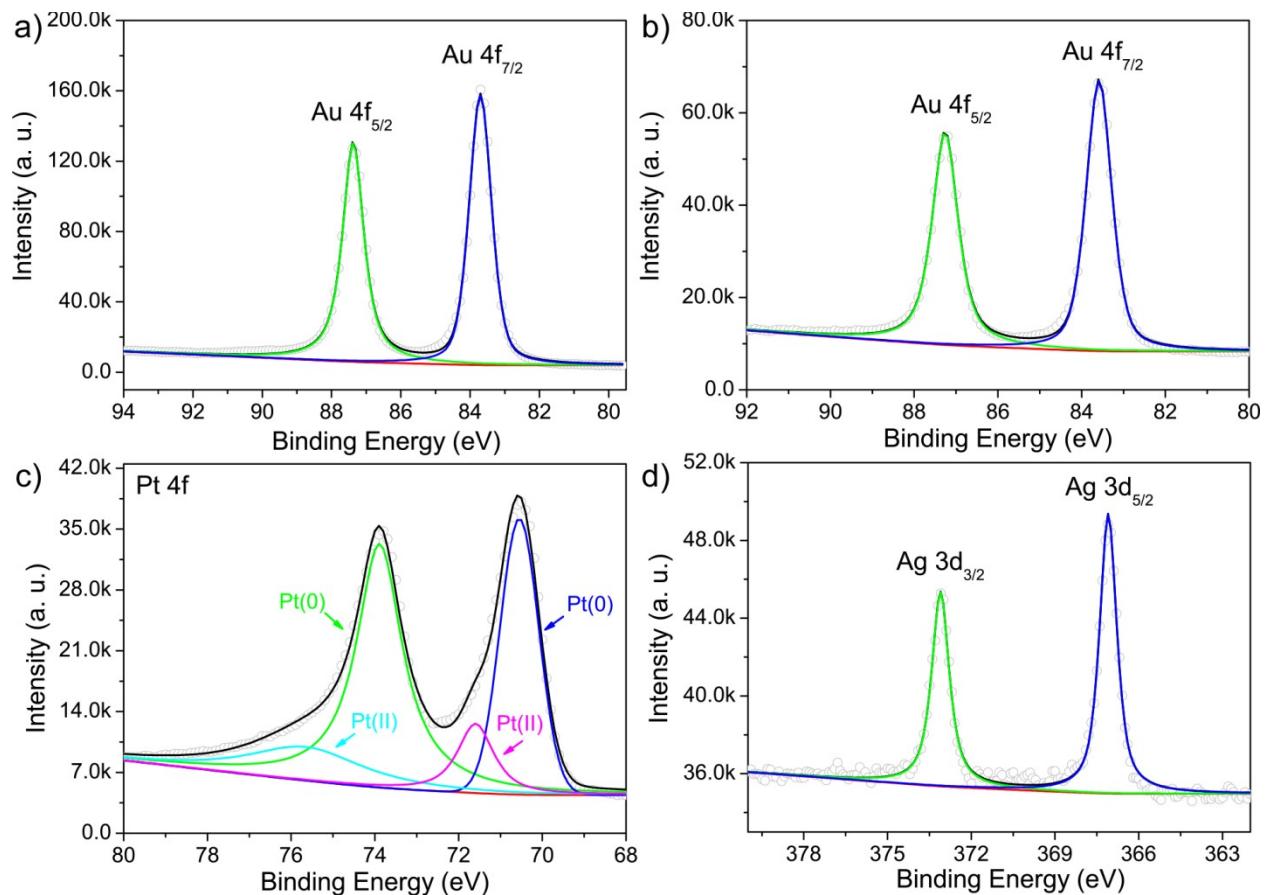
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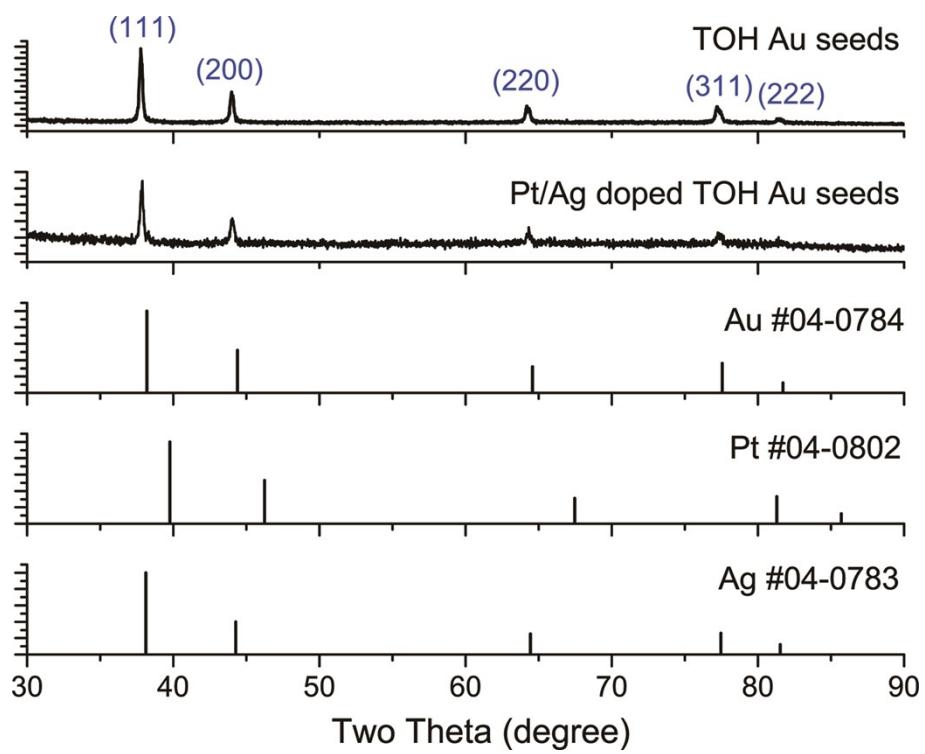
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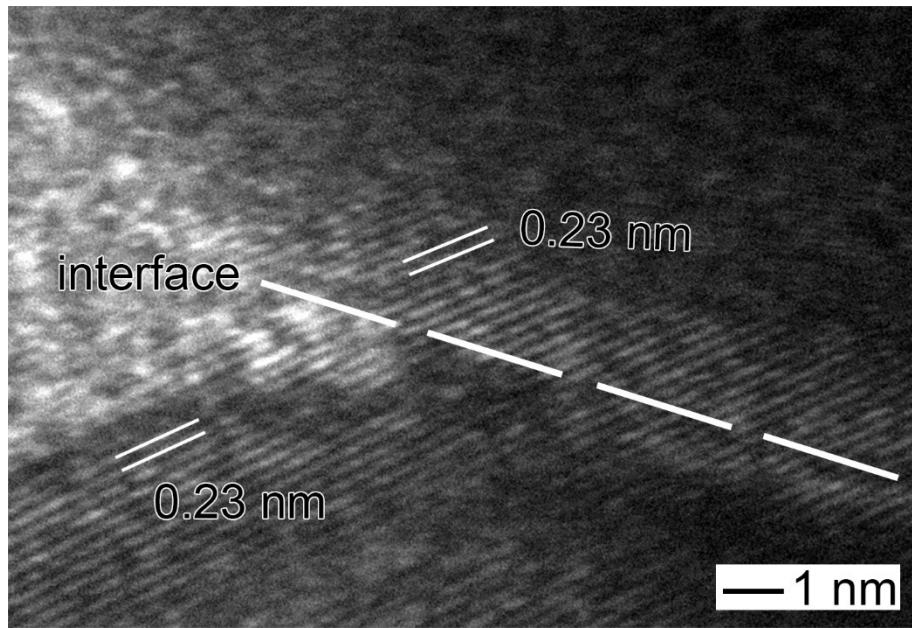
**Figure S1.** Morphology and structural characterizations of Pt/Ag doped TOH Au seeds: a) TEM; b) HAADF-STEM; c-f) EDX-STEM: c) Au+Pt+Ag, d) Ag, e) Pt, f) Au.



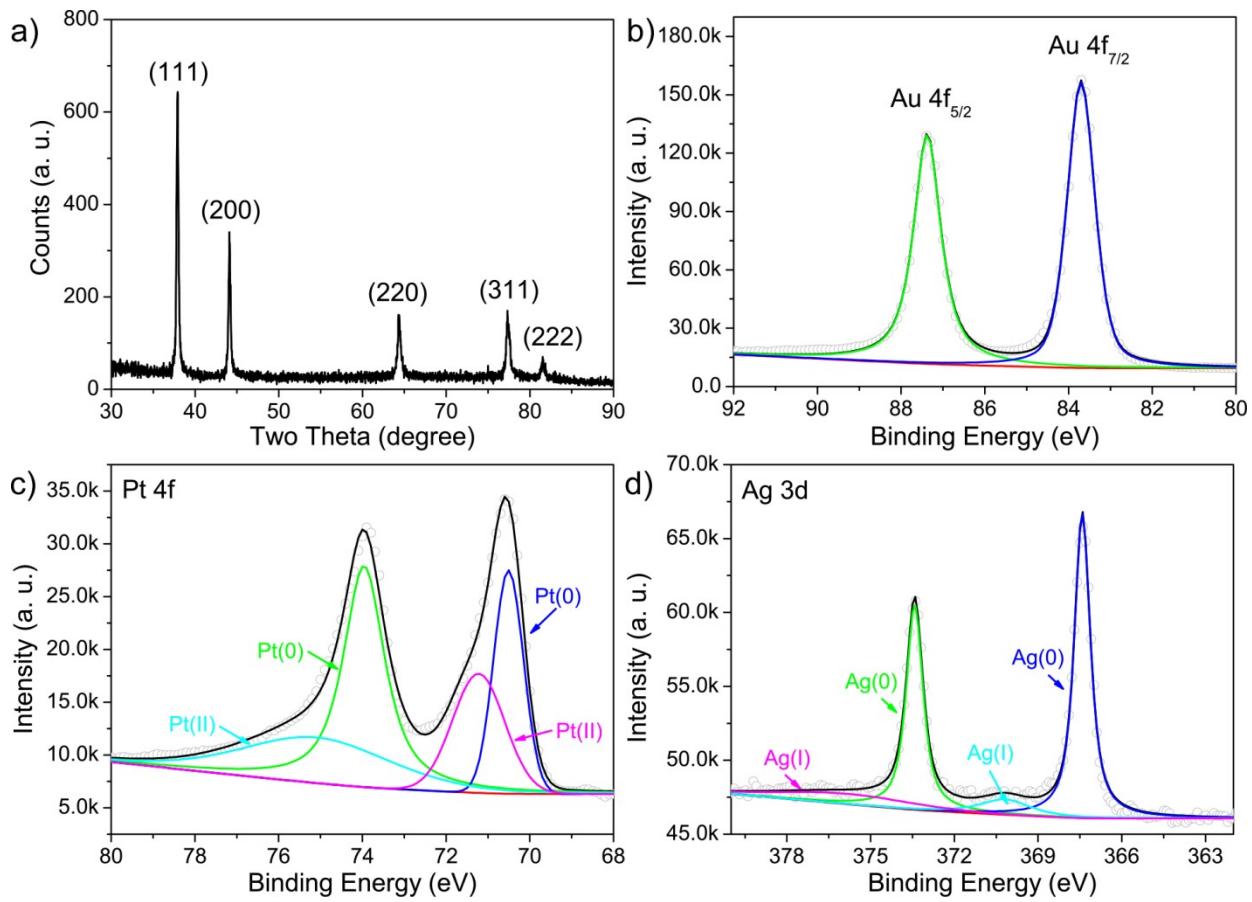
**Figure S2.** High-resolution XPS spectra of a) TOH Au seeds and b-d) Pt/Ag doped TOH Au seeds:  
 a, b) Au 4f; c) Pt 4f; d) Ag 3d.



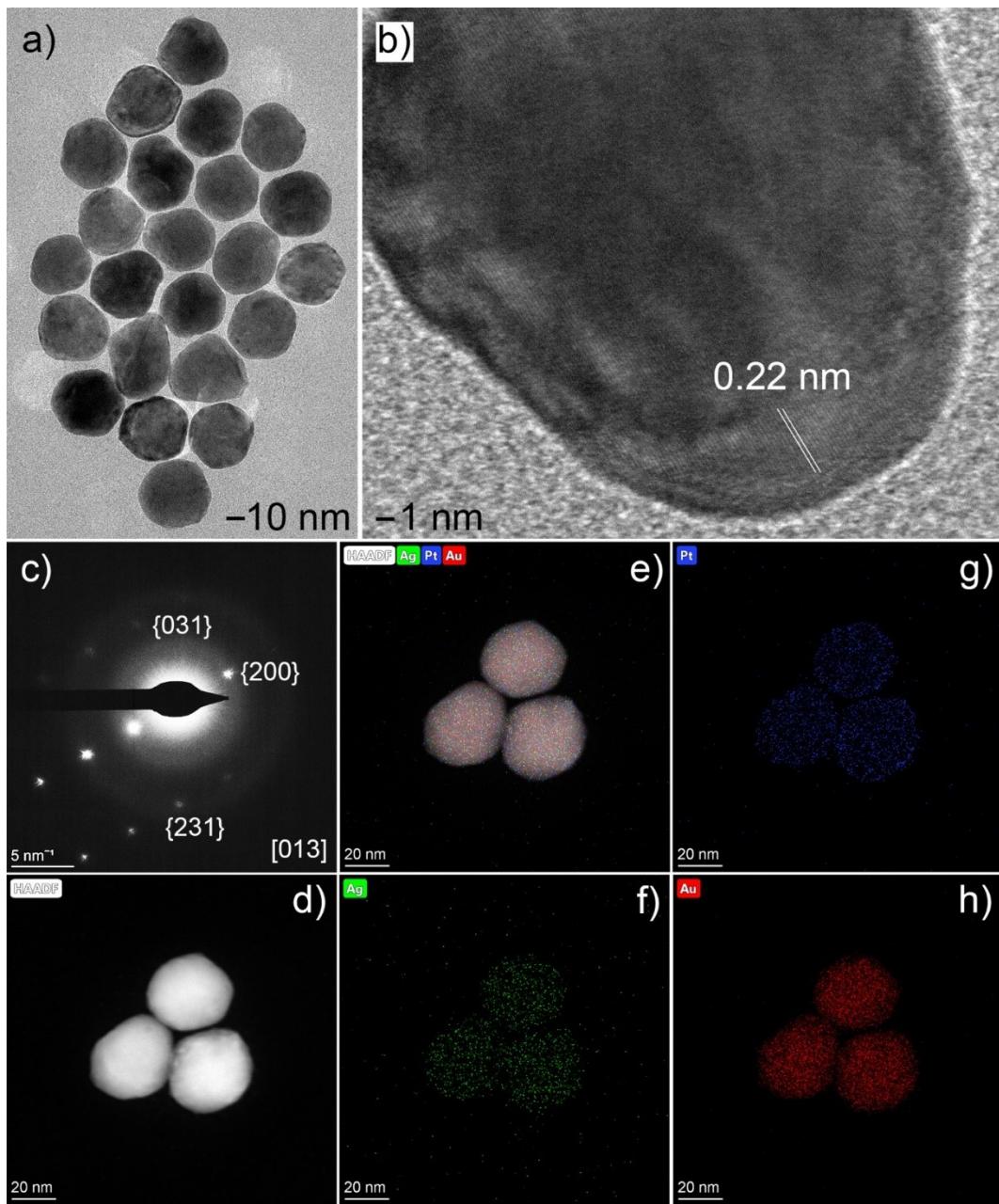
**Figure S3.** XRD patterns of TOH Au seeds and Pt/Ag doped TOH Au seeds.



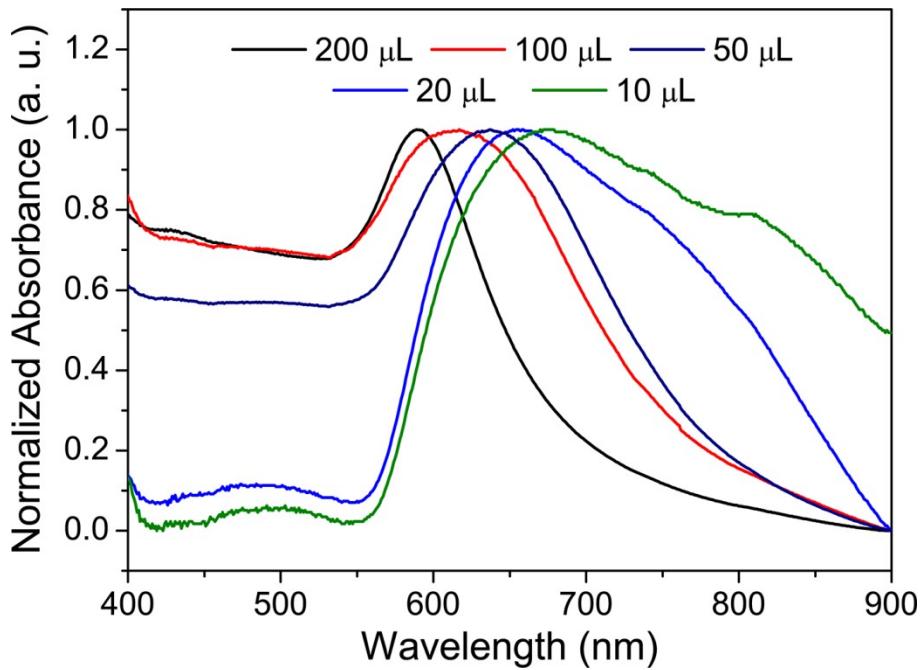
**Figure S4.** HRTEM image of the interface region of an individual Au-based NHJ.



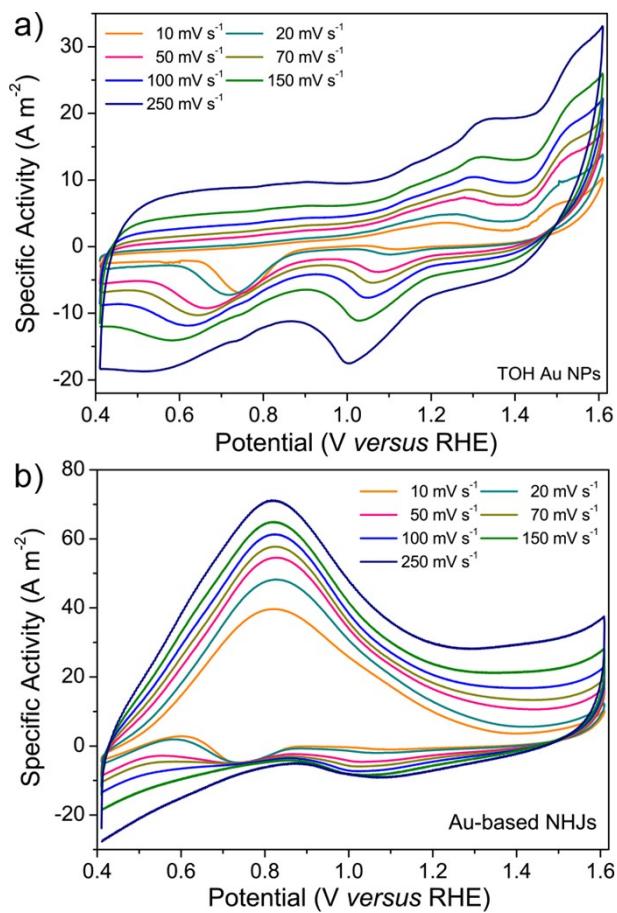
**Figure S5.** a) XRD patterns and b-d) high resolution XPS spectra of Au-based NHJs: b) Au 4f, c) Pt 4f, and d) Ag 3d.



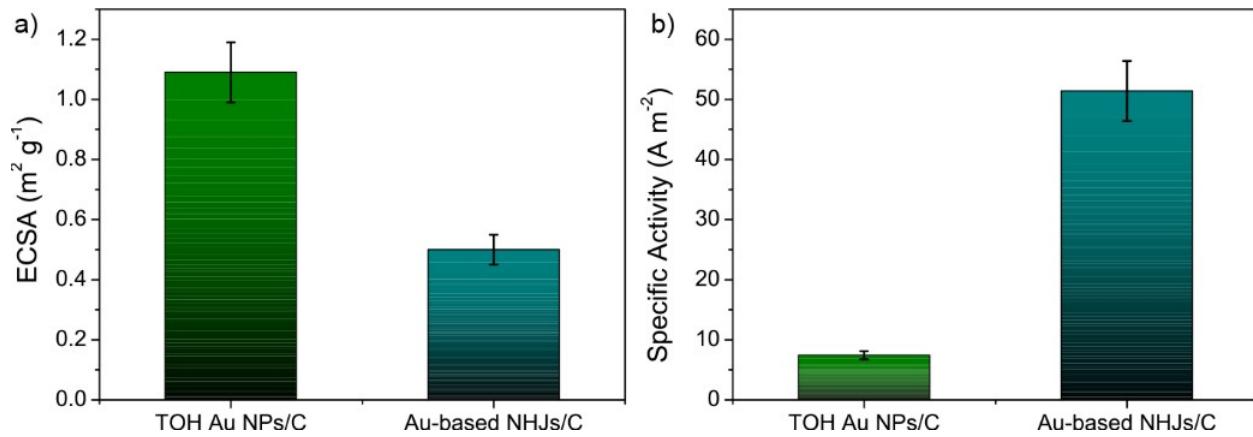
**Figure S6.** Morphology and structural characterizations of products shown in Figure 4d: a) TEM; b) HRTEM; c) SAED; d) HAADF-STEM; e-h) EDX-STEM: e) Au+Pt+Ag, f) Au, g) Pt, h) Au.



**Figure S7.** UV-vis extinction spectra of Au-based products obtained via the standard procedure, except for the use of 10-nm Au seeds stock solution at different volumes.



**Figure S8.** CV traces of a) TOH Au NPs/C and b) Au-based NHJs/C recorded in 0.5 M KOH+1 M methanol under different scan rates.



**Figure S9.** Histograms showing the MOR data of a) ECSA and b) specific activity, which were collected *via* multiple parallel tests under the same measurement conditions as described in the experimental section.

**Table S1.** XRD peak positions of TOH Au seeds, Pt/Ag doped TOH Au seeds, Au-based NHJs.

Sample	Diffraction Peak Position				
	(111)	(200)	(220)	(311)	(222)
TOH Au seeds	38.2	44.4	64.6	77.6	81.5
Pt/Ag doped TOH Au seeds	38.3	44.4	64.7	77.8	/
Au (JCPDS No. 04-0784)	38.2	44.4	64.6	77.5	81.7
Pt (JCPDS No. 04-0802)	39.8	46.2	67.5	81.3	85.7
Ag (JCPDS No. 04-0783)	38.1	44.3	64.4	77.5	81.5

**Table S2.** Statistical size information of Au-based NHJs obtained using different amounts of 10-nm Au seeds.

Sample	volume of Au seeds ( $\mu\text{L}$ )	Part I		Part II	
		d (nm)	$\sigma$ (nm)	d (nm)	$\sigma$ (nm)
Figure 4a	10	90.8	20.6	65.4	13.4
Figure 2	20	79.7	28.4	58.7	21.4
Figure 4b	50	67.8	27.2	44.9	15.3
Figure 4c	100	58.2	8.2	30.8	7.5

**Table S3.** LSPR peak positions of Au-based NHJs with different sizes.

Sample	LSPR Peak Position	
Au-based NHJs (10 $\mu$ L)	676 nm	497 nm
Au-based NHJs (20 $\mu$ L)	654 nm	486 nm
Au-based NHJs (50 $\mu$ L)		637 nm
Au-based NHJs (100 $\mu$ L)		614 nm

**Table S4.** Summary of MOR electrocatalytic performance for Au-based electrocatalysts in the present study.

Electrocatalyst	$E_s^*$ (mV)	$E_p^*$ (mV)	ECSA ( $m^2 g^{-1}$ )	Specific Activity ( $A m^{-2}$ )	$j(t=3000s)$ ( $A m^{-2}$ )
TOH Au NPs	479	1280	1.09	7.42	0.10
Au-based NHJs	435	825	0.500	51.4	21.7

\* $E_s$ =onset potential vs. RHE;  $E_p$ =peak potential vs. RHE.

**Table S5.** Comparison of MOR electrocatalytic performance of Au-based electrocatalysts among recent studies.

Electrocatalyst	Electrolyte	Scan Rate	Electrochemical Activity	Ref.
Trisoctahedron Au nanocrystals	0.5 M KOH+1 M CH <sub>3</sub> OH	20 mV s <sup>-1</sup>	0.139 mA cm <sup>-2</sup>	<sup>1</sup>
Au micromeshes/PDMS	0.5 M KOH+1 M CH <sub>3</sub> OH	20 mV s <sup>-1</sup>	0.264 mA cm <sup>-2</sup>	<sup>2</sup>
Dendritic Au	0.1 M KOH+1 M CH <sub>3</sub> OH	10 mV s <sup>-1</sup>	0.095 mA cm <sup>-2</sup>	<sup>3</sup>
Nanoporous Au	0.5 M KOH+1 M CH <sub>3</sub> OH	20 mV s <sup>-1</sup>	0.088 mA cm <sup>-2</sup>	<sup>4</sup>
Dendritic Au	0.1 M KOH+2 M CH <sub>3</sub> OH	10 mV s <sup>-1</sup>	0.056 mA cm <sup>-2</sup>	<sup>5</sup>
Dealloyed nanosponge Au particles	0.5 M KOH+1 M CH <sub>3</sub> OH	5 mV s <sup>-1</sup>	0.182 mA cm <sup>-2</sup>	<sup>6</sup>
Au-based NHJs	0.5 M KOH+1 M CH <sub>3</sub> OH	20 mV s <sup>-1</sup>	5.14 mA cm <sup>-2</sup>	current work

## References

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