Supplementary Information

Low temperature scalable synthetic approach enabling high bifunctional electrocatalytic performance of $NiCo_2S_4$ and $CuCo_2S_4$ thiospinels

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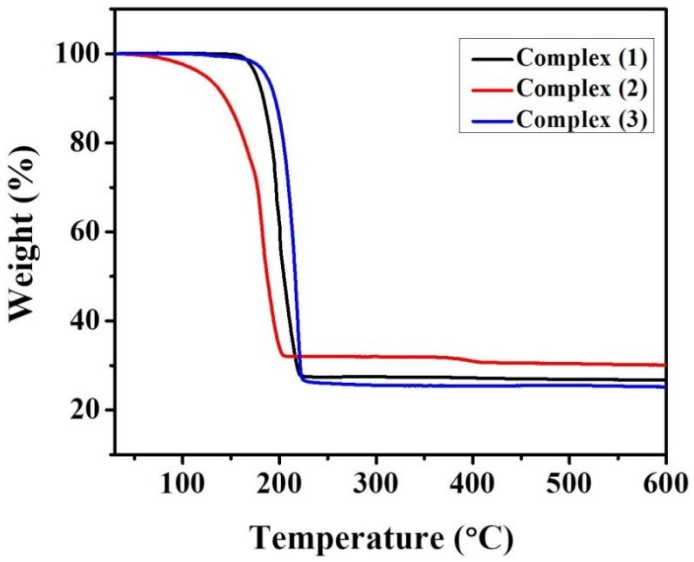

Figure S1. TGA curves of complexes nickel ethyl xanthate (**1**), copper ethyl xanthate (**2**), and cobalt ethyl xanthate (**3**).

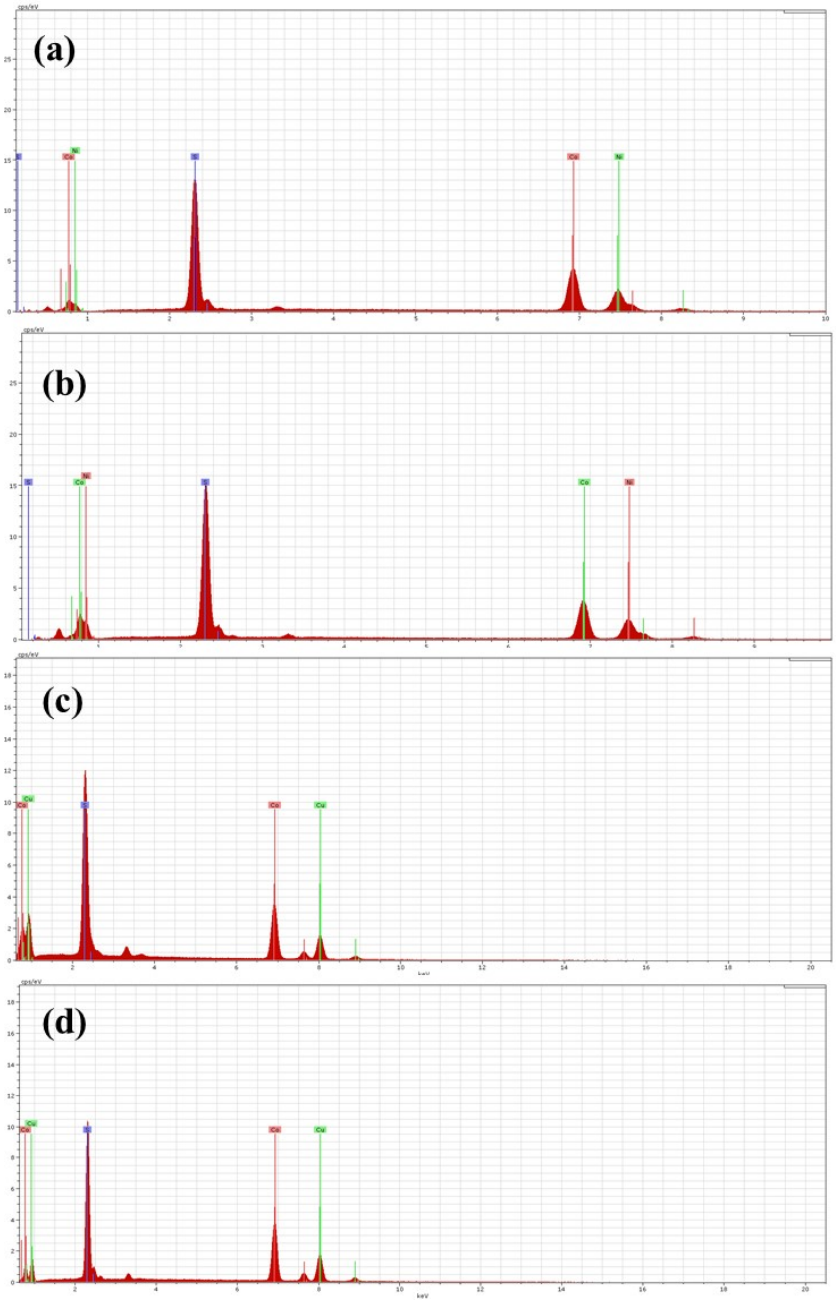

Figure S2. EDX spectra of $NiCo_2S_4$ synthesized at (a) 200 °C and (b) 300 °C. EDX spectra of $CuCo_2S_4$ synthesized at (c) 200 °C and (d) 300 °C

Table S1. Comparison chart with previously reported supercapacitor electrodes.

Electrode Material	Synthetic Method	Specific Capacitance (F/g)	Current Density (A/g)	Reference
$NiCo_2S_4$	Sacrificial template method	437	1	[1]
$NiCo_2S_4$	Solvothermal	519.51	2	[2]
$NiCo_2S_4$	Hydrothermal	744	1	[3]
$NiCo_2S_4$@MnO_2	Hydrothermal	520.7	1	[4]
AMC@ $NiCo_2S_4$	Hydrothermal	651.1	0.6	[5]
$CuCo_2S_4$/polyacrylonitrile		385	1	[6]
$CuCo_2S_4$@$NiCo_2S_4$	Hydrothermal	539.2 C/g	1	[7]
$CuCo_2S_4$	Hydrothermal and sulfuration process	373.4	1	[8]
$CuCo_2S_4$	Anion-exchange and post-annealing	424	1	[9]
$CuCo_2S_4$/Graphene	Hydrothermal	525.4	1	[10]
$CuCo_2S_4$	Two-step hydrothermal approach	~300	0.17 (mA/cm^2)	[11]
$CuCo_2O_4$	Hydrothermal	285.5	0.5	[12]
Co_3O_4	Hydrothermal	272.86	0.5	[13]
$NiCo_2O_4$	Hydrothermal	225	1	[14]
$CoFe_2O_4$	Electrodeposition	768	0.5	[15]
$CuCo_2S_4$	hydrothermal and sulfuration process	373.4	1	[8]
$AgBiS_2$	Melt method	175	0.5	[16]
$CuCo_2S_4$ (CCS-300)	**Solventless approach**	**475**	**0.5**	**This work**
$NiCo_2S_4$ (NCS-300)	**Solventless approach**	**1200**	**0.5**	

*AMC-Activated multiporous carbon

Table S2. Comparison chart with previously reported electrocatalyst electrodes.

Catalyst Materials	Synthetic Method	$\eta_{HER\ (mV)}$ at 10 mA/cm^2	$\eta_{OER\ (mV)}$ at 10 mA/cm^2	Reference
$CuCo_2S_4$/$NiCo_2S_4$	Hydrothermal	206	271	[17]
$CuCo_2S_4$	Hydrothermal	-	310	[18]
$CuCo_2S_4$	Hydrothermal	158	290 (20 mA/cm^2)	[12]
$NiCo_2O_4$	Hydrothermal	-	346	[19]
$NiCo_2S_4$	Solvothermal	226	-	[20]
$NiCo_2S_4$	Hydrothermal followed by sulfidation	-	309	[19]
$NiCo_2O_4$/NF	Solvothermal	~250	~350	[21]

$NiCo_2Se_4$	Hydrothermal		270	[19]
$NiCo_2S_4$/NF	Solvothermal	~200	~330	[21]
Ni_3S_2/NF	Hydrothermal and thermal sulfurization process	271	-	[22]
$NiCo_{-LDH}$/NF	Hydrothermal	231	-	[22]
$NiCo_2S_4$/NF	Hydrothermal	210	260	[23]
$NiCo_2S_4$/NF	Two-step hydrothermal method	191	<251 (40 mA/cm^2)	[24]
$NiCo_2S_4$@N/S-rGO	Solvothermal	-	470	[25]
$NiCo_2O_4$ NS/CC	Hydrothermal	-	368	[26]
$NiCo_2S_4$ NS/CC	Hydrothermal	-	316	[26]
$CuCo_2S_4$	Solution-based chemical route	-	395	[27]
FeO@$CuCo_2S_4$	Hydrothermal	-	240	[28]
CCS-300	**Solventless approach**	**224**	**269**	**This Work**
NCS-300	**Solventless approach**	**209**	**318**	

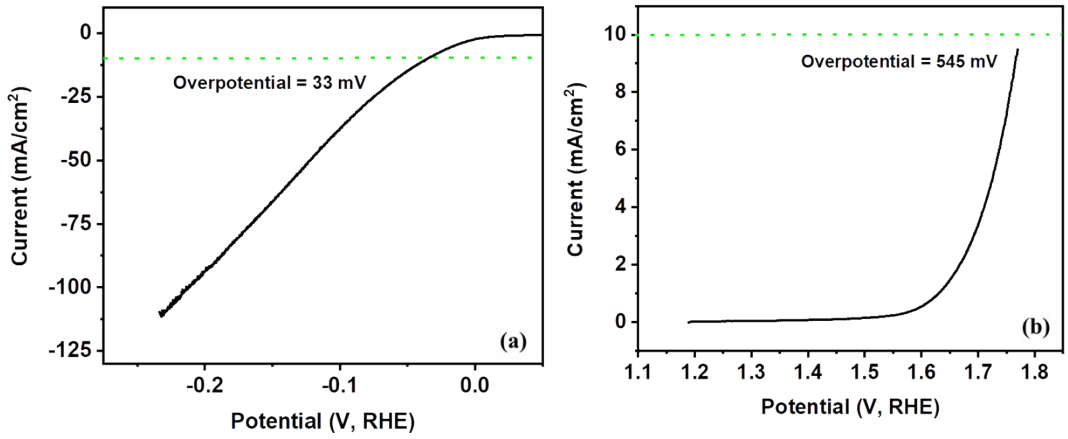

Figure S3. Electrocatalytic activity of commercial platinum for HER (a) and OER (b) processes.

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