Electronic Supplementary Information

Electrocatalysts composed of Co(acetylacetonate)$_2$ molecule and refluxed graphene oxide for oxygen reduction reaction

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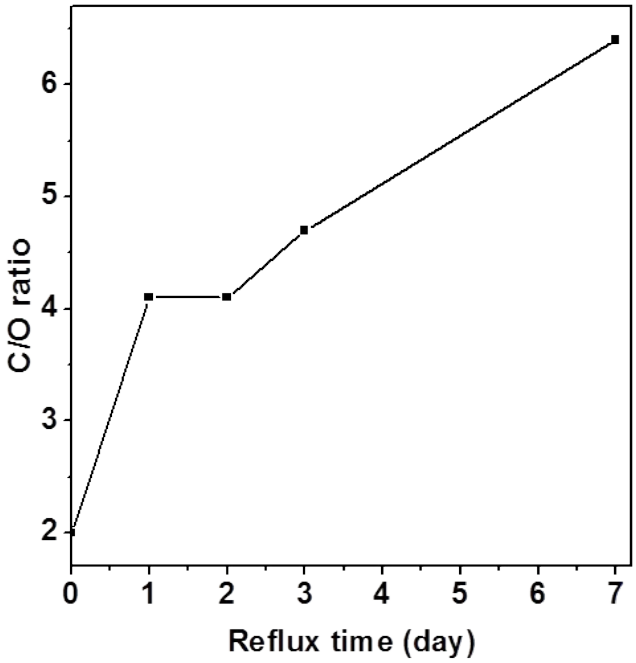

Fig. S1 C/O atomic ratio of Re-G-O samples as a variation of reflux time measured by XPS.

Table S1 EA and XPS results for graphite oxide and Re-G-O materials.

Sample	**EA**			**XPS**		
	C [at%]	O [at%]	C/O ratio	C [at%]	O [at%]	C/O ratio
Graphite oxide	47.4	38.2	1.7	64.2	31.7	2.0
Re-G-O-1	59.9	36.8	2.1	80.5	19.5	4.1
Re-G-O-2	58.4	30.7	2.5	80.3	19.8	4.1
Re-G-O-3	65.4	25.3	3.4	82.5	17.5	4.7
Re-G-O-7	73.3	17.7	5.5	86.5	13.5	6.4

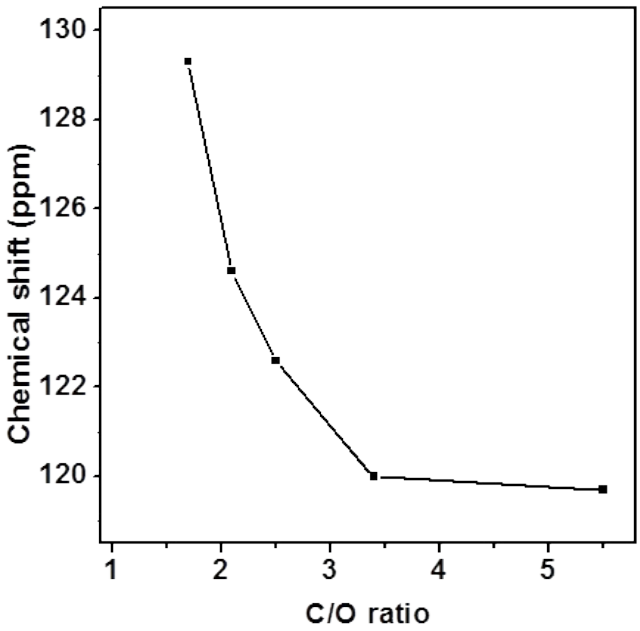

Fig. S2 Relationship between C/O atomic ratio and the chemical shift of sp^2 carbons in ^{13}C MAS SSNMR.

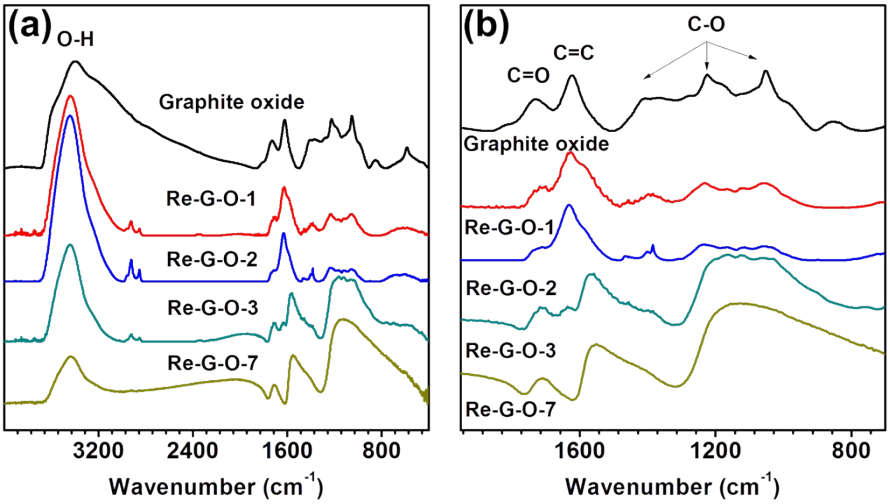

Fig. S3 FT-IR spectra of graphite oxide and a series of Re-G-O materials.

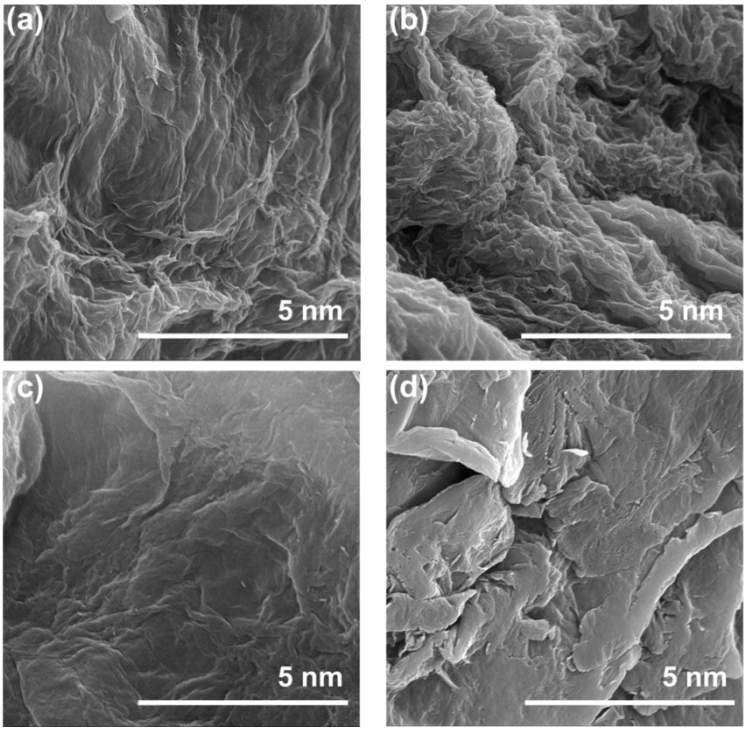

Fig. S4 SEM images of (a) **Re-G-O-1**, (b) **Co(II)-Re-G-O-1**, (c) **Re-G-O-3**, and (d) **Co(II)-Re-G-O-3**.

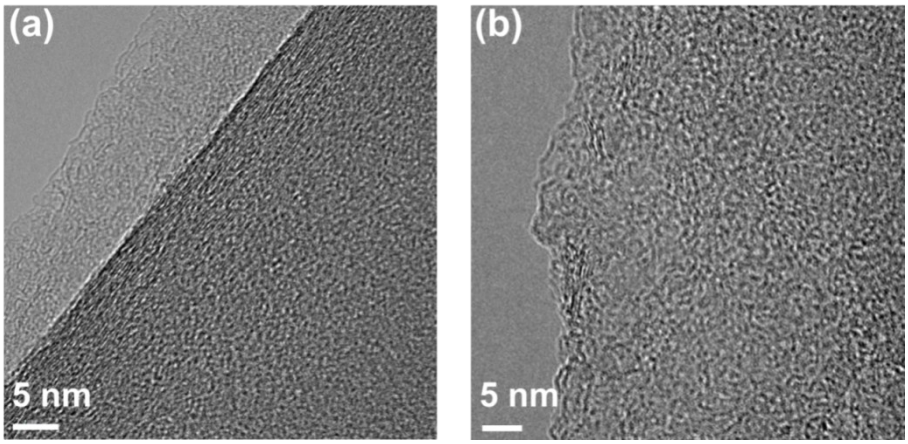

Fig. S5 TEM images of (a) **Re-G-O-3** and (b) **Co(II)-Re-G-O-3**.

Table S2 XPS data for Re-G-O and Co(II)-Re-G-O-based materials, and amounts of Co(II) atoms in Co(II)-Re-G-O samples measured by EDX and EDS, respectively.

Sample	**XPS**				**EDX**	**EDS**
	C [at%]	O [at%]	Co(II) [at%]	C/O ratio	Co(II) [at%]	Co(II) [at%]
Re-G-O-1	59.9	36.8		2.1		
Re-G-O-2	58.4	30.7		2.5		
Re-G-O-3	65.4	25.3		3.4		
Re-G-O-7	73.3	17.7		5.5		
Co(II)-Re-G-O-1	67.9	24.7	4.8	2.8	4.3	
Co(II)-Re-G-O-2	75.2	20.5	2.4	3.7		2.8
Co(II)-Re-G-O-3	80.1	17.0	1.7	4.7	1.7	1.0
Co(II)-Re-G-O-7	83.9	14.4	1.0	5.8		

Table S3 Amounts of each O-containing species calculated from deconvoluted O1*s* XPS spectra.

Sample	O content [at%]	O-1[a] [at%]	O-2[b] [at%]	O-3[c] [at%]	C/O ratio
Re-G-O-1	19.5	3.0	14.9	1.6	4.1
Re-G-O-2	19.8	4.1	14.0	1.7	4.1
Re-G-O-3	17.5	3.3	11.8	2.4	4.7
Re-G-O-7	13.5	4.3	6.8	2.4	6.4
Co(II)-Re-G-O-1	24.7	12.5	10.4	1.8	2.8
Co(II)-Re-G-O-2	20.5	9.6	9.2	1.7	3.7
Co(II)-Re-G-O-3	17.0	6.7	9.0	1.3	4.7
Co(II)-Re-G-O-7	14.4	7.7	4.7	2.0	5.8

[a] O-1 is C=O group (531.5 eV), [b] O-2 is C-O group (533.0 eV), [c] O-3 is O-C=O group (533.8 eV).

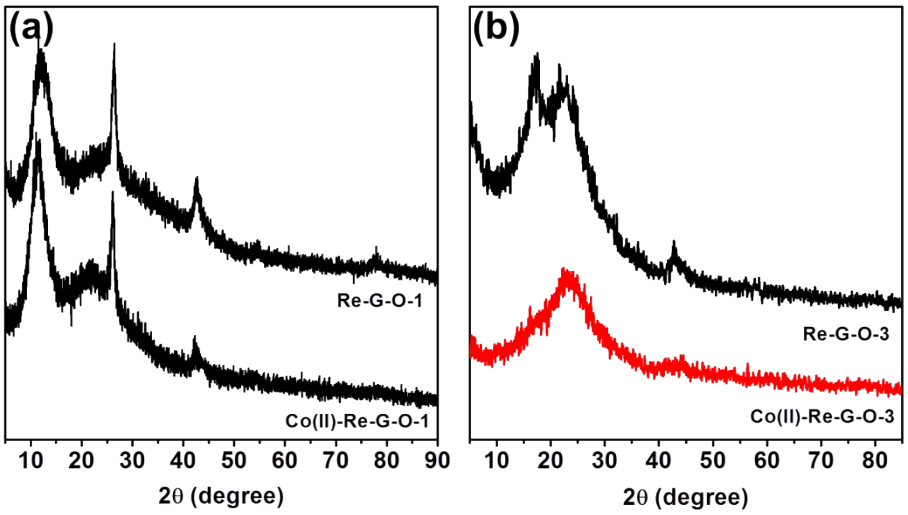

Fig. S6 XRD patterns of (a) **Re-G-O-1** and **Co(II)-Re-G-O-1**, and (b) **Re-G-O-3** and **Co(II)-Re-G-O-3**.

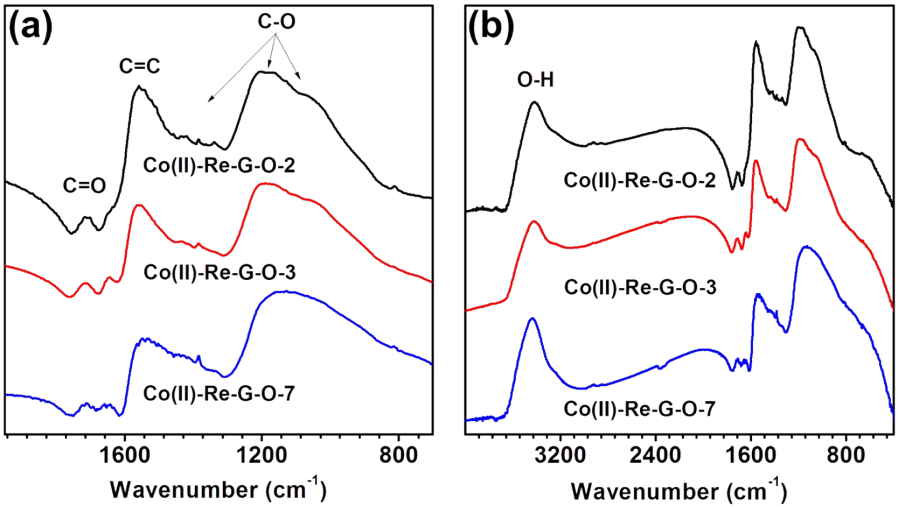

Fig. S7 FT-IR spectra of a set of Co(II)-Re-G-O materials.

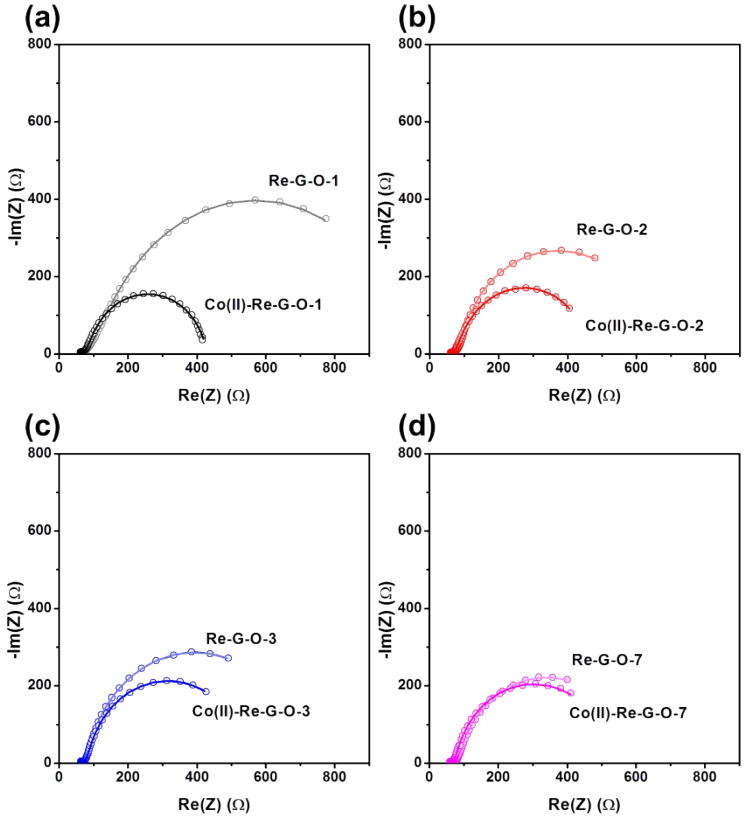

Fig. 8 Electrochemical impedance spectroscopy data for (a) Re-G-O-1 and **Co(II)-Re-G-O-1**, (b) **Re-G-O-2** and **Co(II)-Re-G-O-2**, (c) **Re-G-O-3** and **Co(II)-Re-G-O-3**, and (d) **Re-G-O-7** and **Co(II)-Re-G-O-7** in O_2-saturated 0.1 M KOH at rotating speed of 1600 rpm.

Table S4 Comparison of the amount of atoms measured by XPS and the ORR activity in alkaline media (0.1 M KOH) for a series of Re-G-O and Co(II)-Re-G-O samples.

Sample	**Atomic%**			**Onset potential** [V *vs.* RHE]	**Half-wave potential** [V *vs.* RHE]	**Kinetic current @ 0.75 V** [mA cm^{-2}]	**Limited-current density @ 0.2 V** [mA cm^{-2}]	**Electron transfer number @ 0.2 V**
	C	O	Co(II)					
Re-G-O-1	80.5	19.5		0.82	0.68	-1.17	2.90	**3.0**
Re-G-O-2	80.3	19.8		0.82	0.675	-1.09	2.85	2.8
Re-G-O-3	82.5	17.5		0.83	0.68	-1.19	3.00	2.9
Re-G-O-7	86.5	13.5		0.80	0.665	-0.83	2.62	2.7
Co(II)-Re-G-O-1	67.9	24.7	4.8	0.836	0.69	-1.79	3.88	3.5
Co(II)-Re-G-O-2	75.2	20.5	2.4	0.84	0.70	-2.10	3.90	3.6
Co(II)-Re-G-O-3	80.1	17.0	1.7	0.84	0.71	-1.84	3.81	3.4
Co(II)-Re-G-O-7	83.9	14.4	1.0	0.80	0.68	-1.37	3.49	3.3

Table S5 BET Surface area data of Re-G-O-1~7 and Co(II)-Re-G-O-1~7 samples.

Sample	**BET surface area** [m^2/g]	**Sample**	**BET surface area** [m^2/g]
Re-G-O-1	394.3	Co(II)-Re-G-O-1	50.0
Re-G-O-2	331.3	Co(II)-Re-G-O-2	163.7
Re-G-O-3	380.1	Co(II)-Re-G-O-3	112.7
Re-G-O-7	369.7	Co(II)-Re-G-O-7	102.5

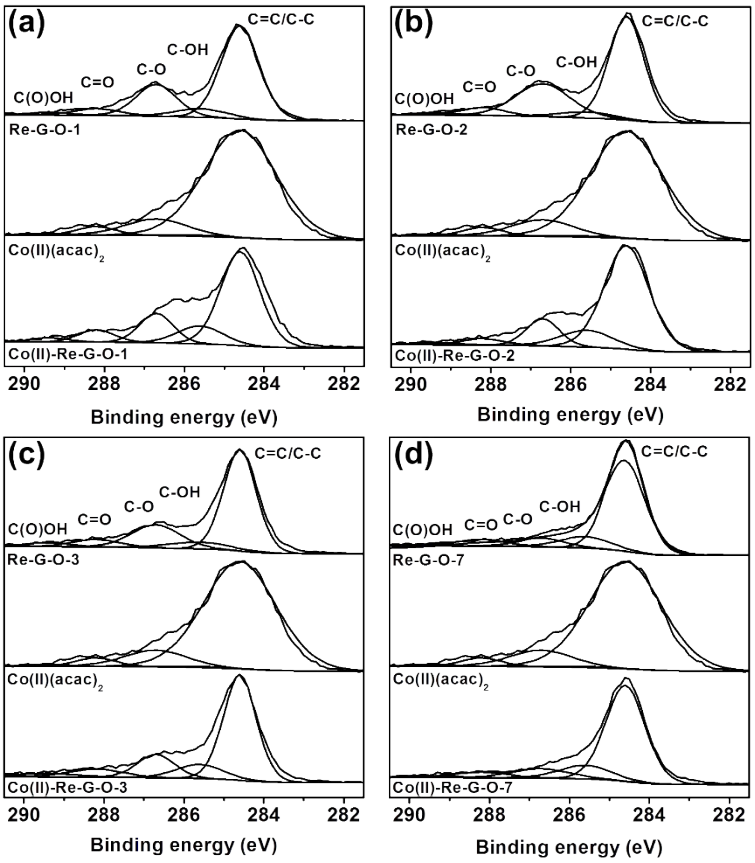

Fig. S9 Deconvoluted C1*s* XPS spectra of Re-G-O, Co(II)(acac)$_2$, and Co(II)-Re-G-O samples.

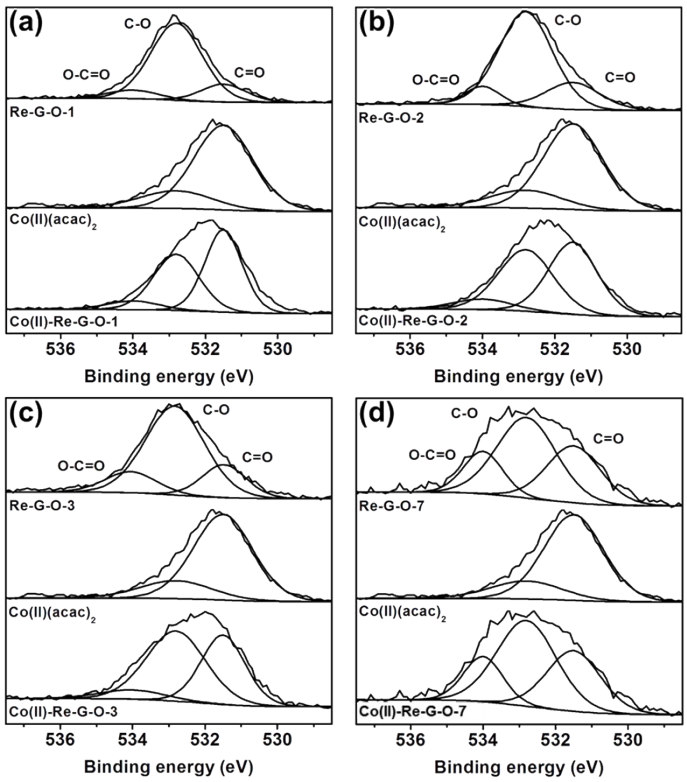

Fig. S10 Deconvoluted O1*s* XPS spectra of Re-G-O, Co(II)$(acac)_2$, and Co(II)-Re-G-O samples.

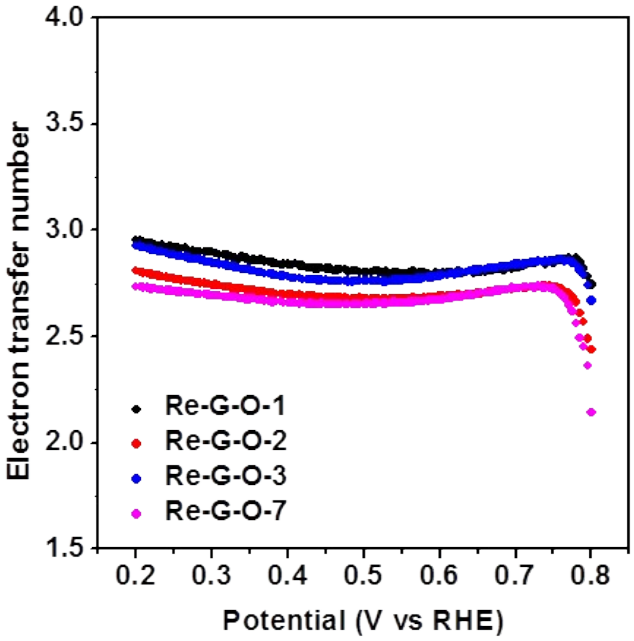

Fig. S11 Electron transfer numbers for a set of Re-G-O samples.

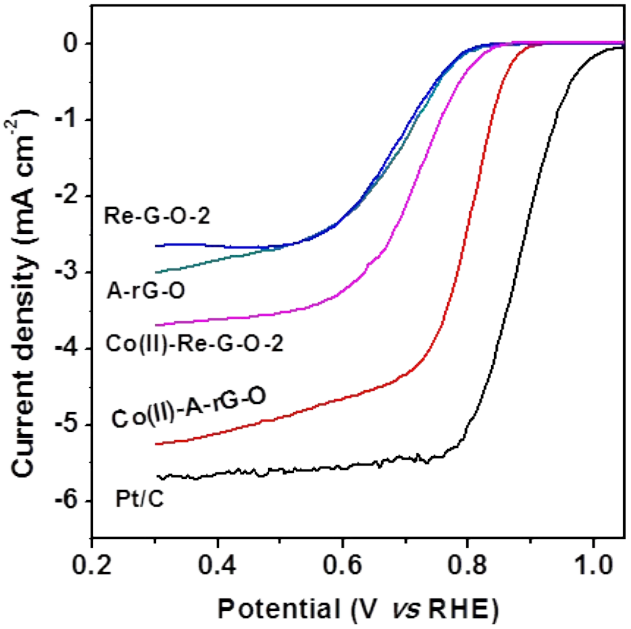

Fig. S12 LSV curves for **Re-G-O-2**, **A-rG-O**, **Co(II)-Re-G-O**, **Co(II)-A-rG-O**, and Pt/C catalyst in O_2-saturated 0.1 M KOH at a rotating speed of 1600 rpm.