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Mechanochemically Synthetized PAN-Based Co-N-Doped Carbon Materials as Electrocatalyst for Oxygen Evolution Reaction

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Abstract: We report a new class of polyacrylonitrile (PAN)-based Co-N-doped carbon materials that can act as suitable catalyst for oxygen evolution reactions (OER). Different Co loadings were mechanochemically added into post-consumed PAN fibers. Subsequently, the samples were treated at 300 °C under air (PAN-A) or nitrogen (PAN-N) atmosphere to promote simultaneously the Co_3O_4 species and PAN cyclization. The resulting electrocatalysts were fully characterized and analyzed by X-ray diffraction (XRD) and photoelectron spectroscopy (XPS), transmission (TEM) and scanning electron (SEM) microscopies, as well as nitrogen porosimetry. The catalytic performance of the Co-N-doped carbon nanomaterials were tested for OER in alkaline environments. Cobalt-doped PAN-A samples showed worse OER electrocatalytic performance than their homologous PAN-N ones. The PAN-N/3% Co catalyst exhibited the lowest OER overpotential (460 mV) among all the Co-N-doped carbon nanocomposites, reaching 10 mA/cm^2. This work provides in-depth insights on the electrocatalytic performance of metal-doped carbon nanomaterials for OER.

Keywords: mechanochemical synthesis; carbon N-doped; Co_2O_3 nanoparticles; PAN; OER

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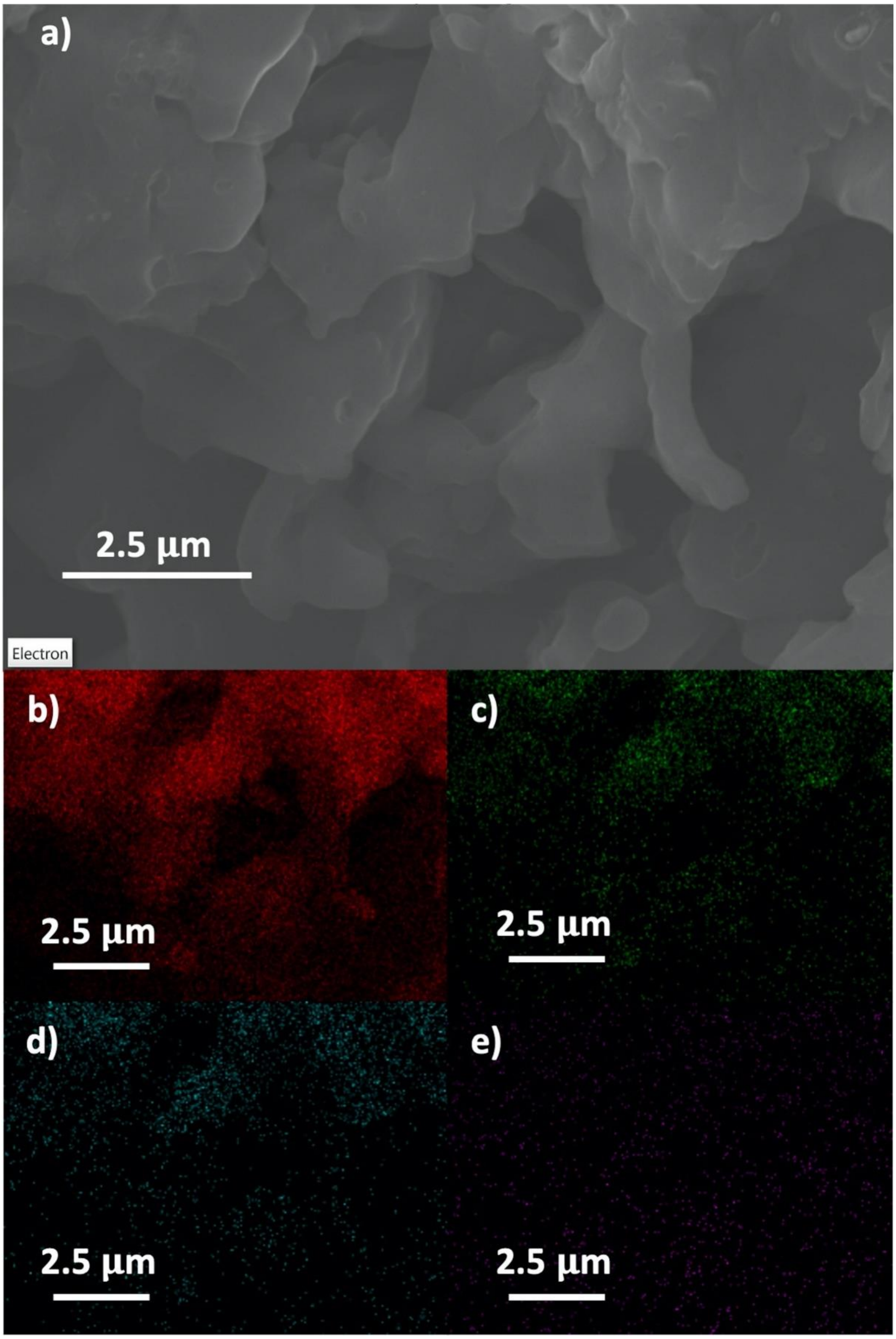

Figure S1. (**a**) SEM-EDX for PAN-N (0% Co) and their elemental mapping of (**b**) carbon, (**c**) nitrogen, (**d**) oxygen and (**e**) Iron, no detection, we can discard the impurity by iron ball milling.

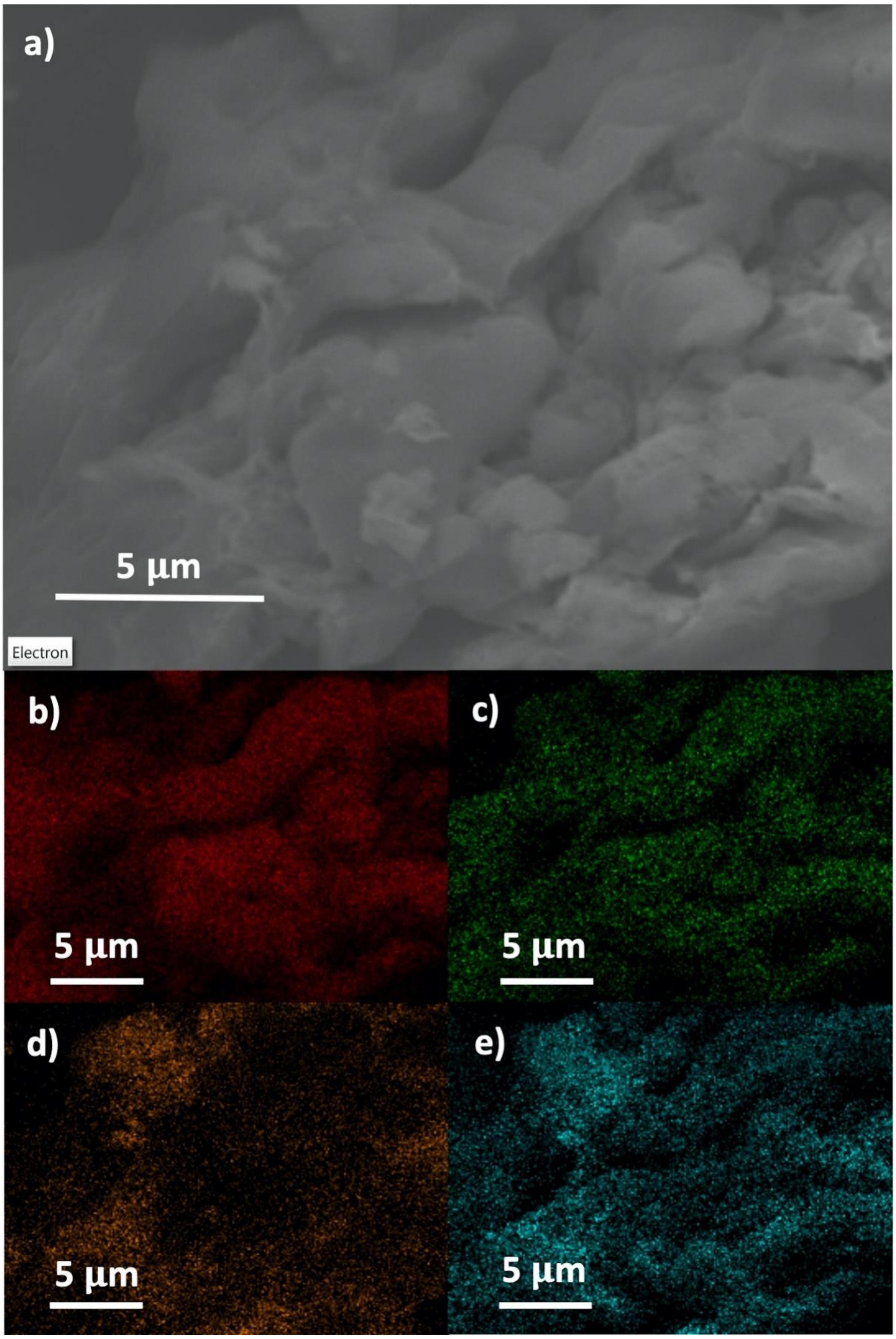

Figure S2. (**a**) SEM-EDX for PAN-AIR/3%Co and their elemental mapping of (**b**) carbon, (**c**) nitrogen, (**d**) cobalt and (**e**) oxygen.

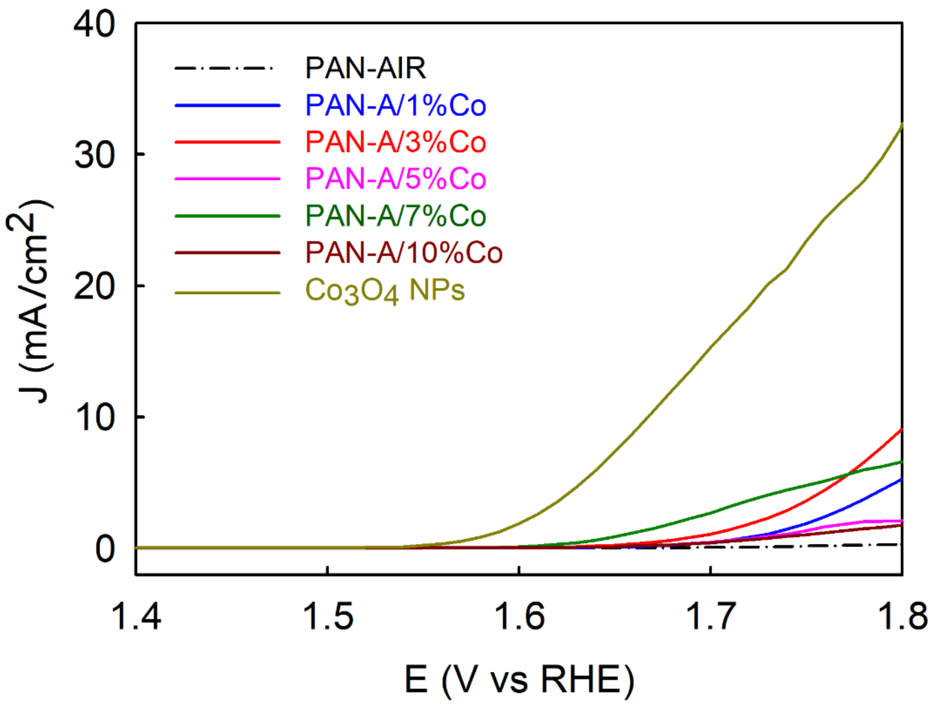

Figure S3. OER polarization curves obtained for the different PAN-AIR/Co samples, including the reference (Co_3O_2 NPs) in 0.5 M KOH solution at 2 mV/s.

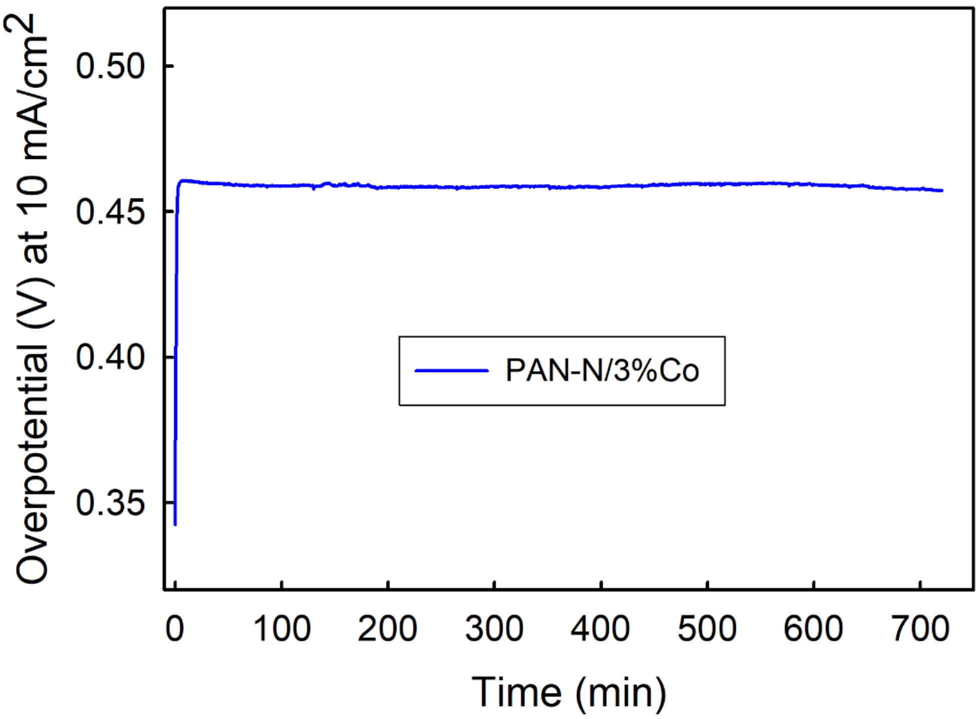

Figure S4. Chronopotentiometry measurements at J = 10 mA/cm² for the PAN-N/3%Co sample.

Table S1. A comparison on the OER electrocatalytic activity of our PAN-N/3%Co sample and other reported Co@Carbon-based OER catalysts.

Co@C Based Catalysts	η_{10} (mV)	Tafel slope (mV/dec)	References
Co_9S_8/C nanosheets	434	122	[S1]
CoCMP	340	87	[S2]
Co@C600	277	46	[S3]
Co–B/C	320	75	[S4]
Co/CoO-C	340	84.5	[S5]
Co/CoO_x@BS-NCNTs-700	360	112	[S6]
CoC_x/FeCo@C/rGO	390	99.1	[S7]
MoS_2/Co–N–CN_2	410	74	[S8]
Co–Ni–P/MoS	300	71	[S9]
Co_3S_4@$NiMoO_4$	320	102	[S10]
Co-Mo_2C@NGCS-2	360	54	[S11]
$ACTP_2$@Co,N-800	374	98	[S12]
$Co_2N_{0.67}$-BHPC	340	90	[S13]
Co/N-CNT (1500 SCCM)	310	66	[S14]
PBA-Pd-Co/C	500		[S15]
PAN-N/3%Co	460	60	This work

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