Electronic Supplementary Material (ESI) for Catalysis Science & Technology.

Supporting Information for:

Enhanced Oxygen Evolution Catalysis by Aluminium-Doped Cobalt Phosphide Through *In Situ* Surface Area Increase

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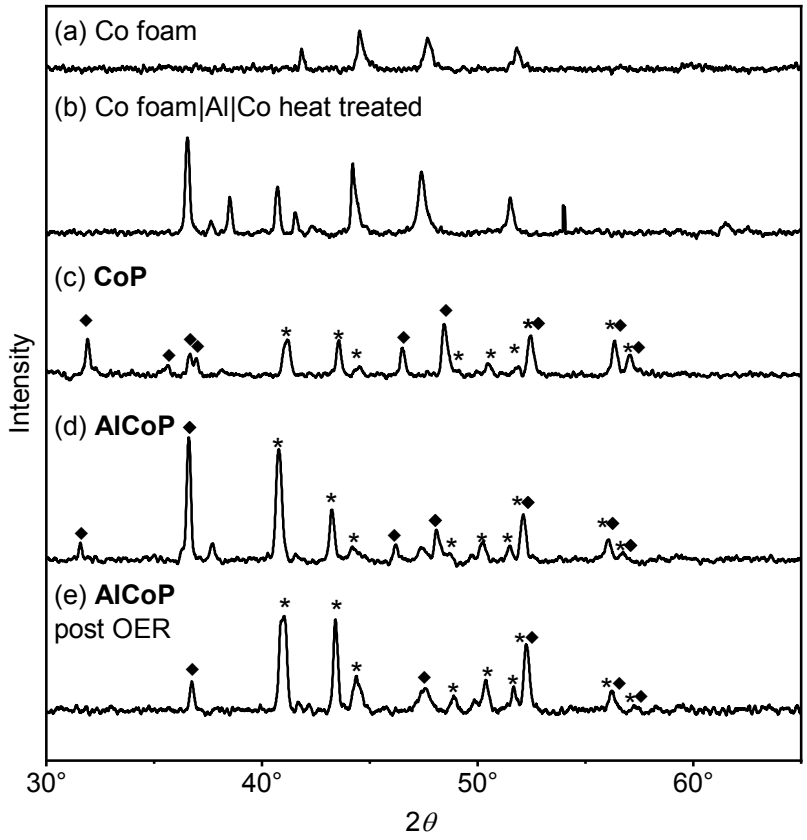

Figure S1 XRD patterns of (a) bare Co foam (phase mixture of hexagonal (ICDD no. 00-005-0727, $P6_3/mmc$) and cubic (ICDD no. 04-006-8067, $Fm\text{–}3m$) Co), (b) foam after Al and Co sputtering and heat treatment at 600 °C (a mixture of pristine hexagonal and cubic Co, as well as monoclinic $Al(OH)_3$ (ICDD no. 04-016-3462, $C2/m$,) and cubic $CoAl_2O_4$ (ICDD no. 04-006-3962, $Fd\text{–}3m$,) as Al-containing phases, from the atmospheric oxidation of metallic Al), (c) phosphorised Co foam **CoP** (d) the synthesised **AlCoP** foam and (e) the **AlCoP** foam after stability testing for 150 h. The ◆ symbol indicates orthorhombic CoP (ICDD no. 03-065-2593, *Pnma* and * indicates orthorhombic Co_2P (ICDD no. 01-070-8359, *Pnma*).

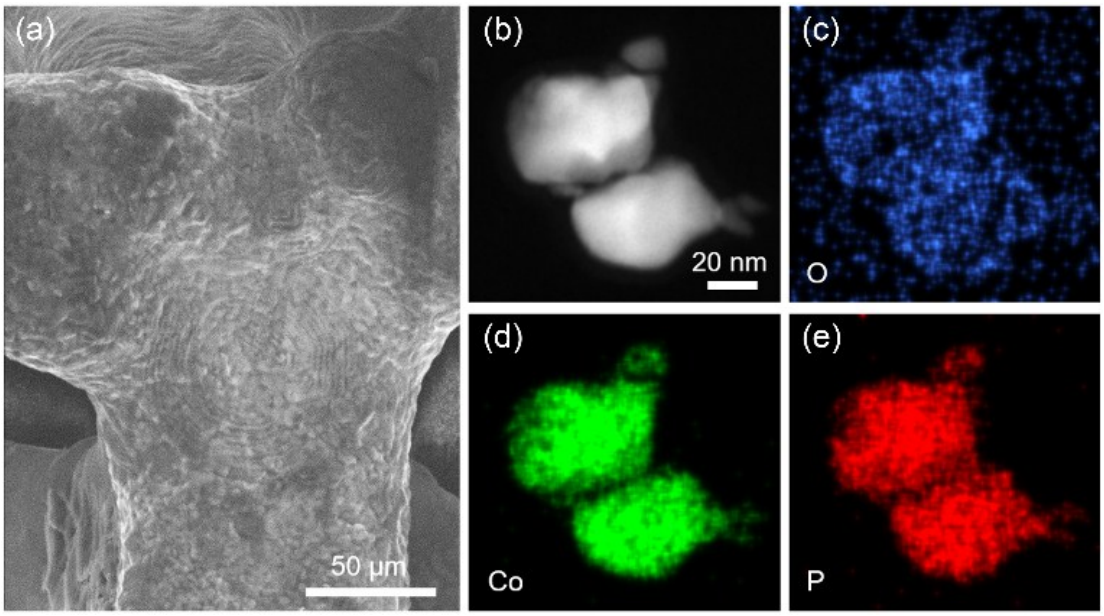

Figure S2 (a) SEM image of as-synthesised **CoP** foam anode. (b) HAADF-STEM image of representative particles. STEM-EDX elemental maps of (c) O, (d) Co and (e) P.

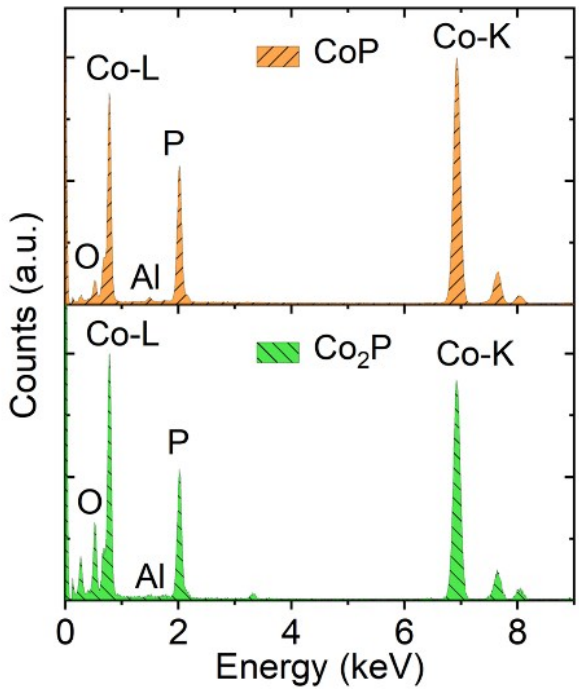

Figure S3 Representative EDX spectra collected from **AlCoP**, confirming the expected CoP and Co_2P phase composition.

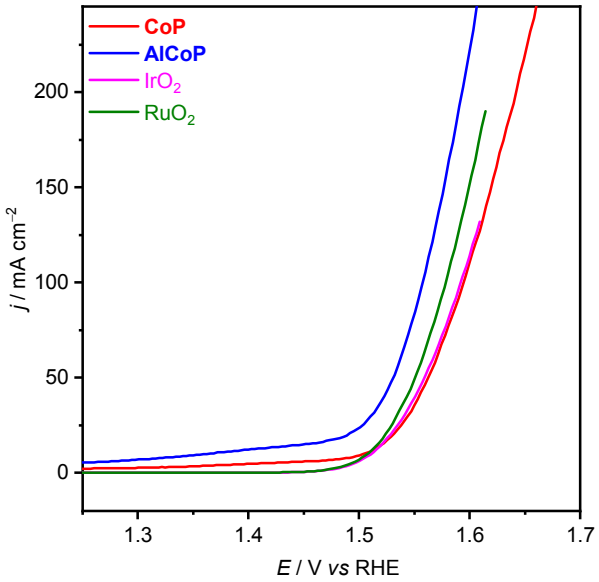

Figure S4 Cyclic voltammetry (5 mV/s) of the **AlCoP** and **CoP** foams in 1.0 M KOH alongside reference data for IrO_2 and RuO_2 measured under the same conditions. The IrO_2 and RuO_2 were deposited on a Ni foam current collector and the data is taken from reference S1 – it should be noted that the surface areas of the reference catalysts may differ from the cobalt-based materials but these data show that the **AlCoP** catalyst performs comparably to typical benchmarks.

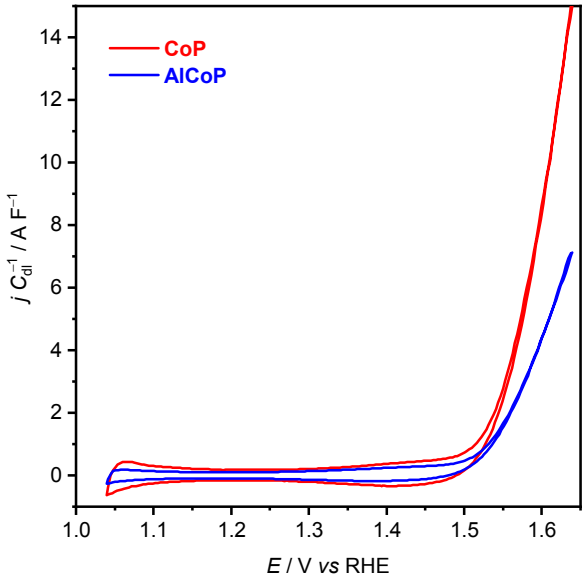

Figure S5 Cyclic voltammetry data presented in Figure 2a, with currents normalised to double layer capacitance calculated in Figure 2c.

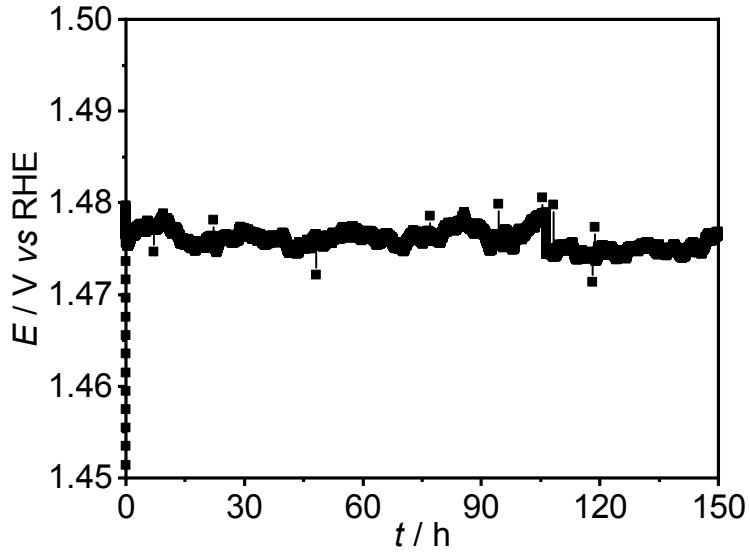

Figure S6 Controlled-current (10 mA cm^{-2}) OER over **AlCoP** foam electrocatalyst conducted in 1.0 M KOH.

(a) **CoP** (b) **AlCoP**

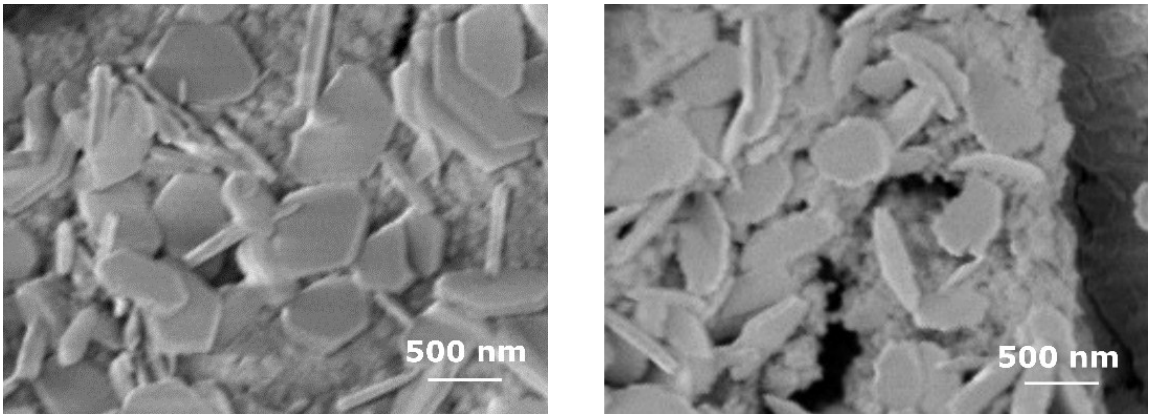

Figure S7 SEM of (a) **CoP** and (b) **AlCoP** after electrochemical OER testing in 1 M KOH (up to 1.45 V *vs*. RHE for 10 mins).

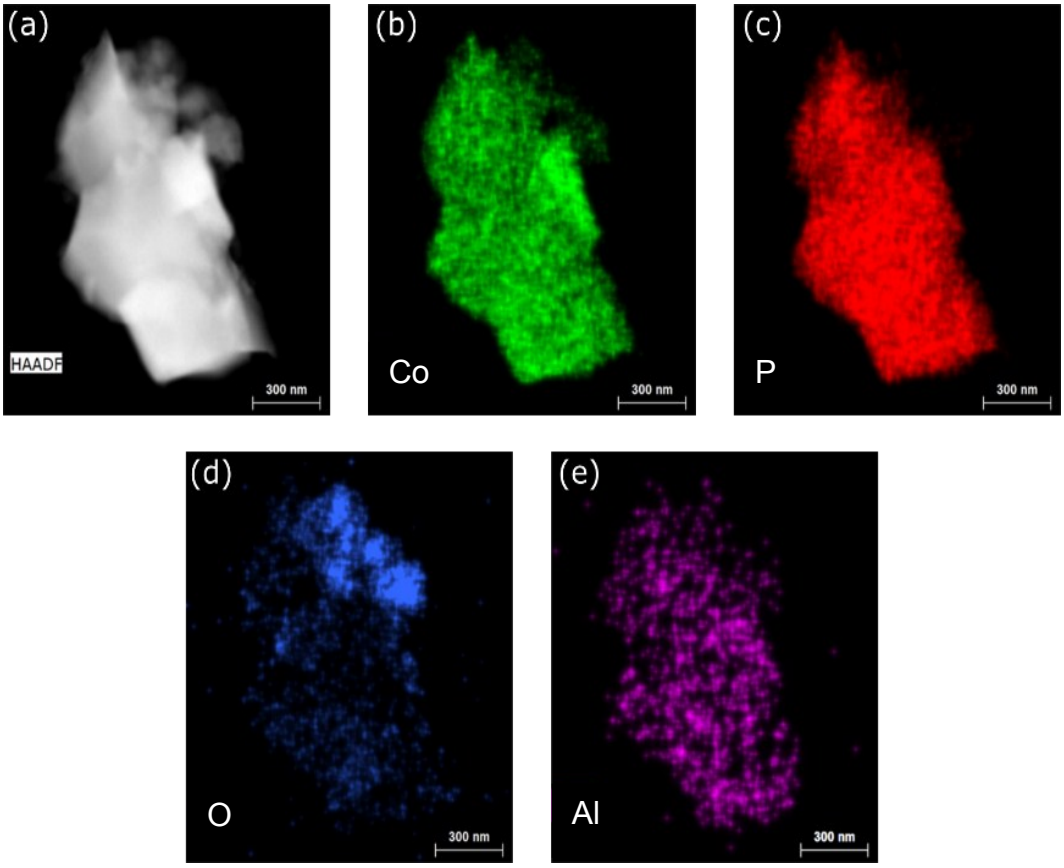

Figure S8 (a) HAADF-STEM image of representative **AlCoP** particle after 150 h stability testing and STEM-EDX elemental maps of (b) Co, (c) P, (d), O and (e) Al.

Table S1 Relative elemental composition values with respect to Co measured by EDX in as-prepared **CoP** and **AlCoP**, and **AlCoP** after stability testing (150 h) including standard deviations of multiple measurements on the same sample.

	CoP	**AlCoP**	**AlCoP** after stability testing
Co	1.0 ± 0.19	1.0 ± 0.10	1.0 ± 0.03
P	1.1 ± 0.08	0.46 ± 0.02	0.30 ± 0.02
Al	0.027 ±0.007	0.17 ± 0.007	0.19 ± 0.01

Table S2 Raman band positions and relative intensity for **CoP** and **AlCoP**

Peak Raman Shift / cm^{-1}		**Band Strength**
CoP	**AlCoP**	
157	153	Strong
180	-	Weak
204	199	Strong
265	261	Strong
282	276	Strong
321	315	Weak
360	354	Strong
451	446	Strong

Supporting references

(S1) D. K. Mann, J. Xu, N. E. Mordvinova, V. Yannello, Y. Ziouani, N. González-Ballesteros, J. P. S. Sousa, O. I. Lebedev, Y. V. Kolen'ko, M. Shatruk, *Chem. Sci.,* **2019**, *10* (9), 2796–2804.