Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

In Situ Synthesis and *In Operando* NMR Studies of High-Performance Ni_5P_4-Nanosheet Anode

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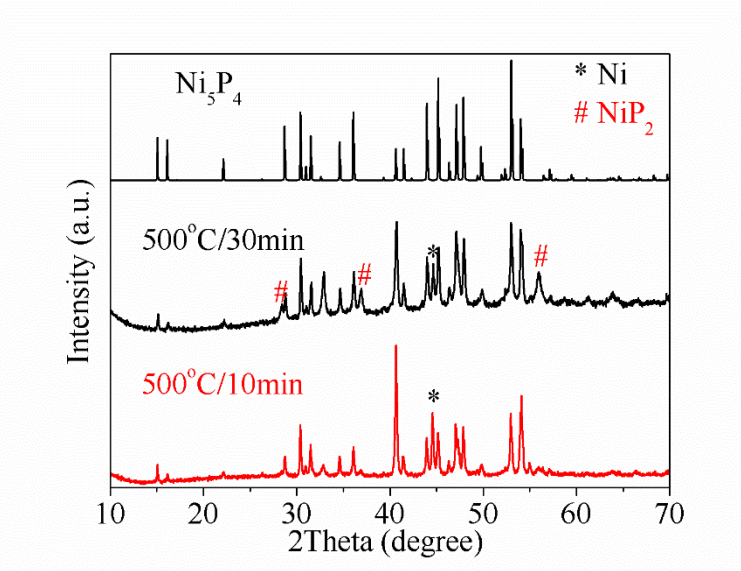

Fig. S1 XRD patterns for the *in situ* synthesized nickel phosphide anodes. Standard PDF files are shown at the top for reference.

The powder X-ray diffraction pattern of the synthesized Ni_5P_4 nanosheets is shown in Fig. S1 along with the XRD patterns simulated for bulk Ni_5P_4. By comparison, these two patterns show resemblance but with difference, which may originate from the effects of particle size and preferred orientations of crystal lattices in the nanosheets. ^{31}P NMR has confirmed that the synthesized nanosheets at 500°C/10min are phase pure and exhibits only the characteristics of local structures as in Ni_5P_4 (Fig. 4).

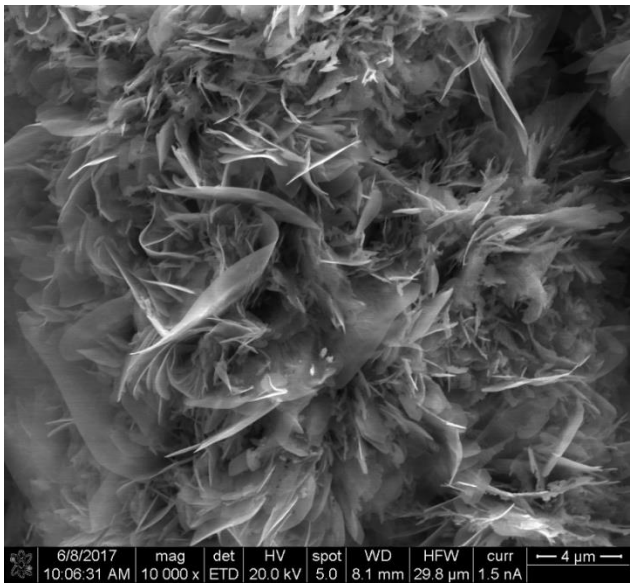

Fig. S2 SEM image of the *in situ* synthesized Ni_5P_4 nanosheets, treated for 30 minutes at 500 ℃.

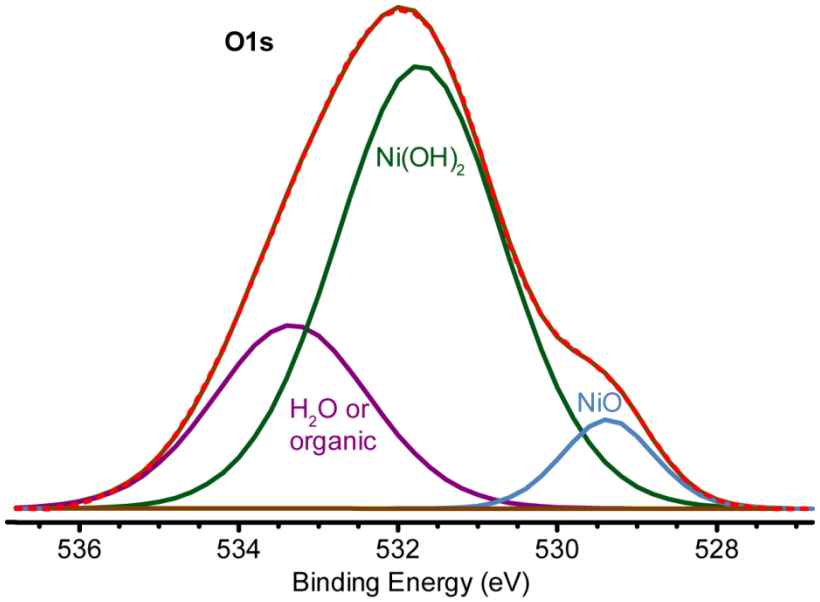

Fig. S3 O_{1s} XPS spectrum and simulation for nickel foam.

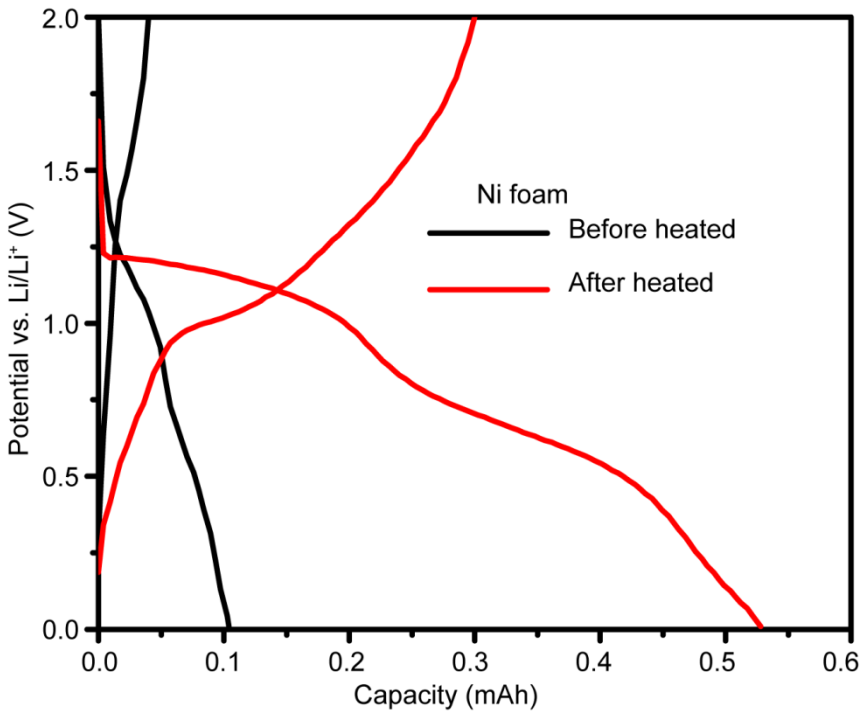

Fig. S4 The 1st-cycle electrochemical profiles of nickel foam before and after heating treatment.

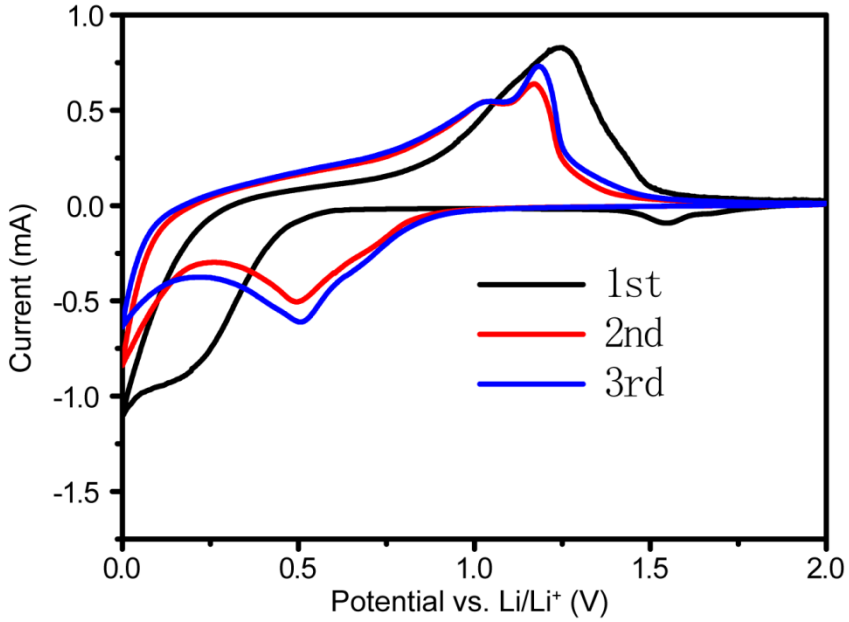

Fig. S5 Cyclic Voltammetry of Ni_5P_4//Li cells for the 1st three cycles at a scan rate of 0.2 mV/s.

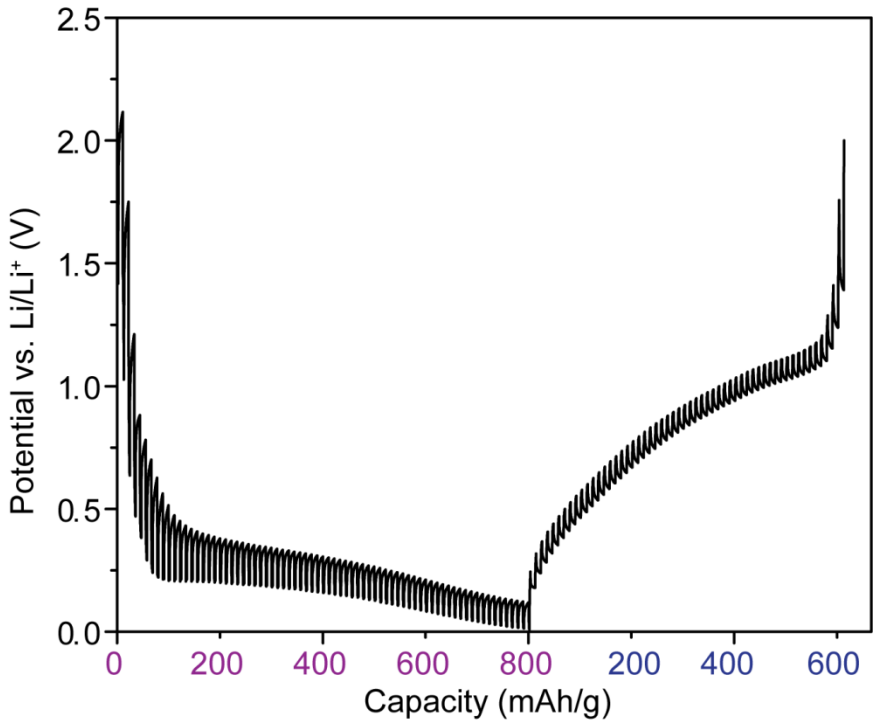

Fig. S6 Galvanostatic Intermittent Titration Technique (GITT) plot of a Ni_5P_4/Li half-cell battery during the 1st cycle.

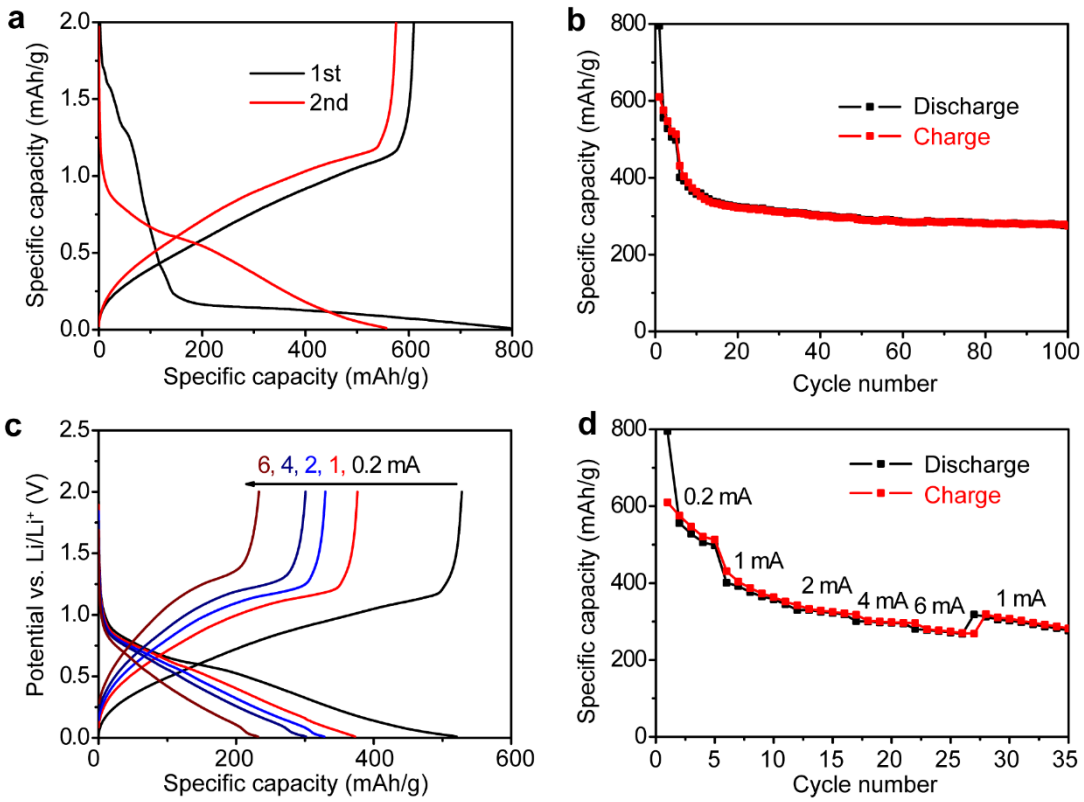

Fig. S7 Electrochemical performance of the Ni_5P_4 anode treated for 30 min in Ni_5P_4/Li half cells. a) discharge-charge profile for the first 2 cycles, and b) the corresponding capacities as a function of cycle number at constant applied current. c) and d) are the electrochemical profiles and capacities obtained at various currents.

Tab. S1 Fitting results of AC impedance spectra in Fig. 2b using the equivalent circuit model

	R_s (Ω)	R_{ct} (Ω)	R_{SEI} (Ω)	R_{total} (Ω)	CPE1-T (F)	CPE1-P	CPE2-T (F)	CPE2-P
Pristine	5.01	134.7		139.7	$2.5*10^{-5}$	0.74		
1^{st} cycle	4.95	1.43	4.35	10.73	$5.5*10^{-7}$	1.06	$3.8*10^{-4}$	0.68
100^{th} cycle	3.64	1.56	12.55	17.75	$7.6*10^{-6}$	0.91	$2.1*10^{-4}$	0.72