

## Supplemental Information

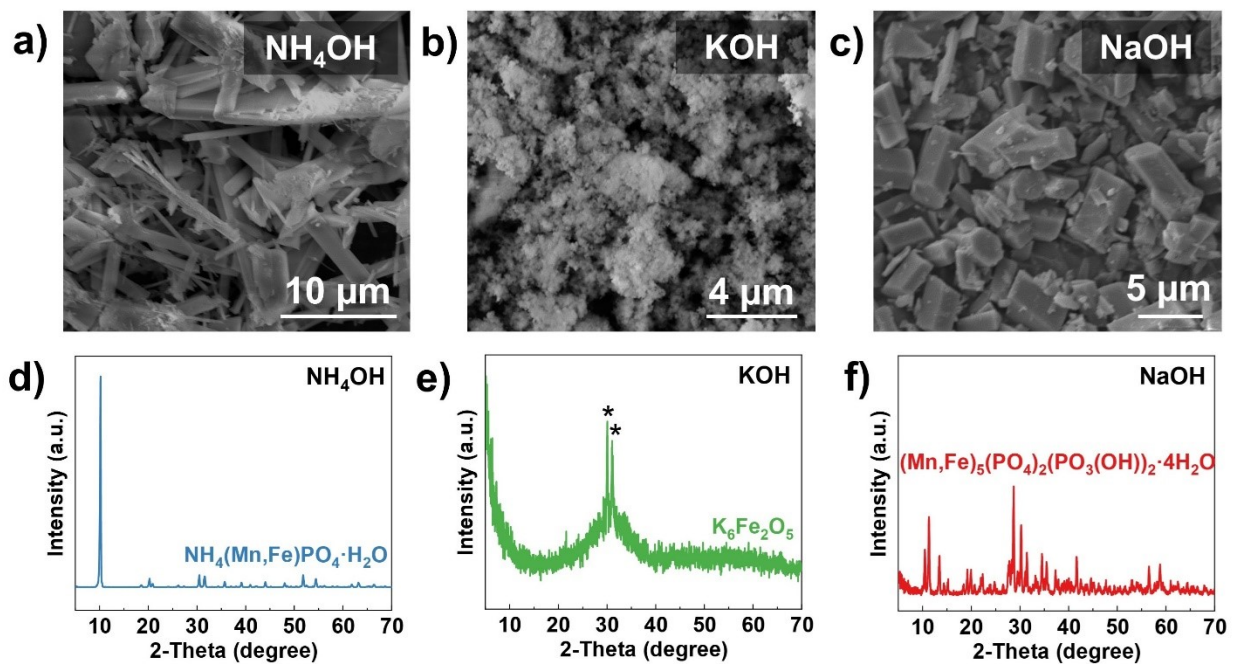
### **Ammonia-free synthesis of lithium manganese iron phosphate cathodes via a co-precipitation reaction**

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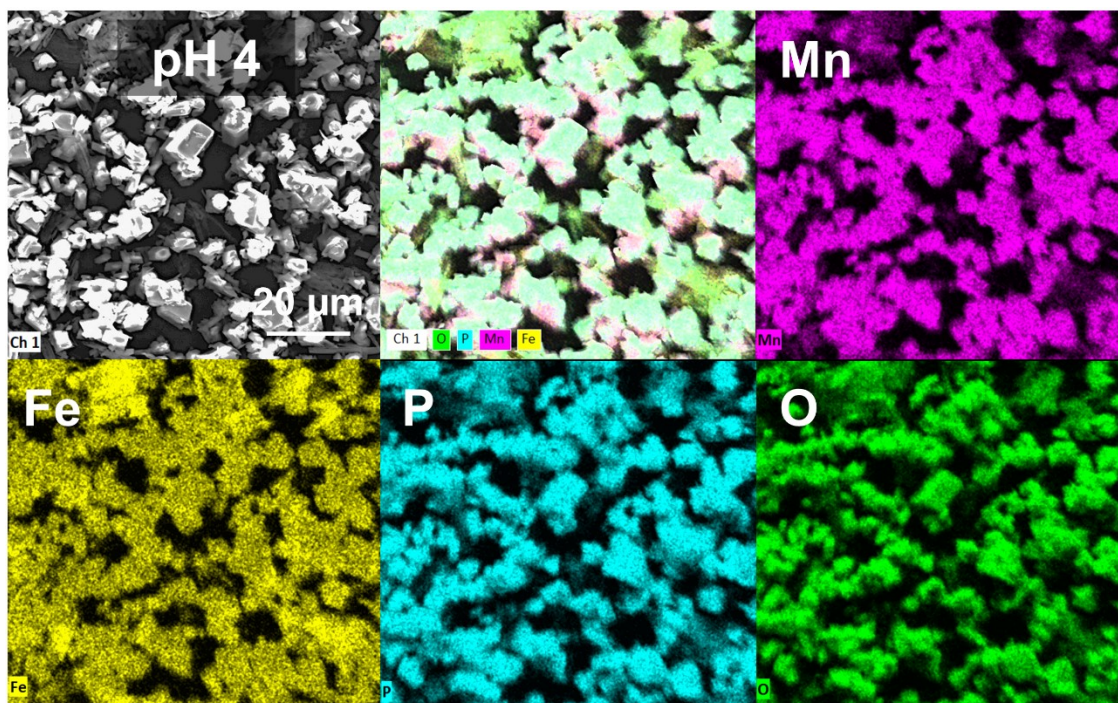
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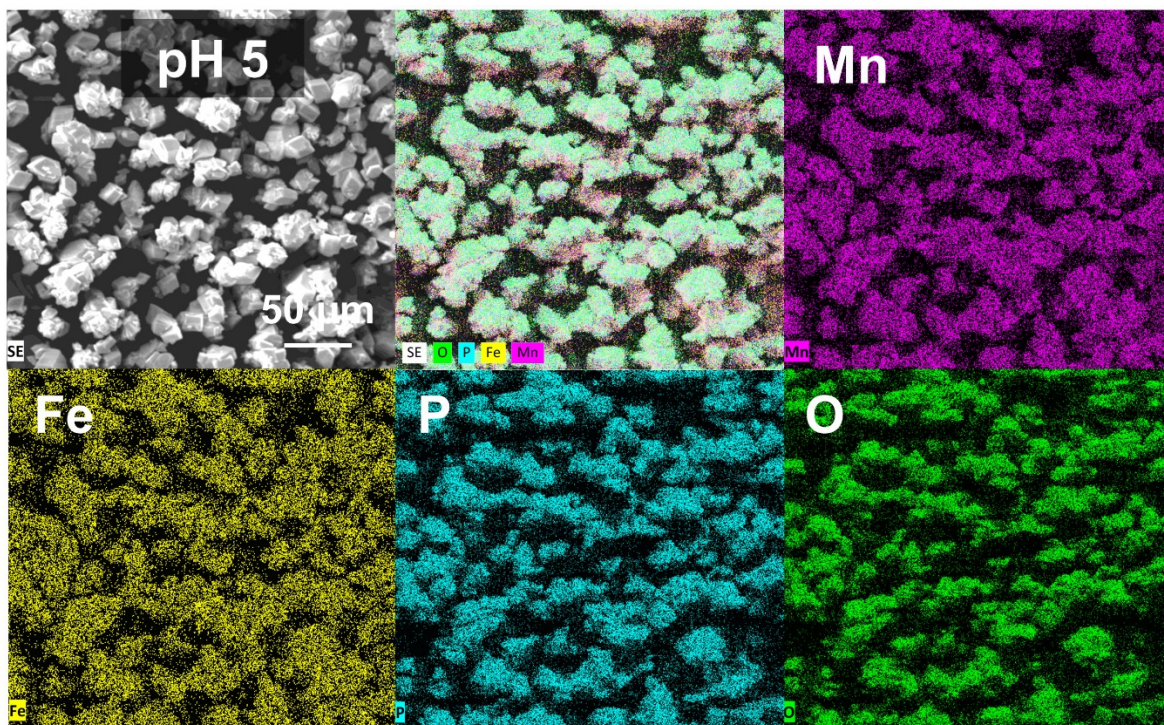


**Fig. S1** Morphological and structural analysis of the precursors obtained with different basic solutions, including (a, d)  $\text{NH}_4\text{OH}$ , (b, e)  $\text{KOH}$ , and (c, f)  $\text{NaOH}$ .

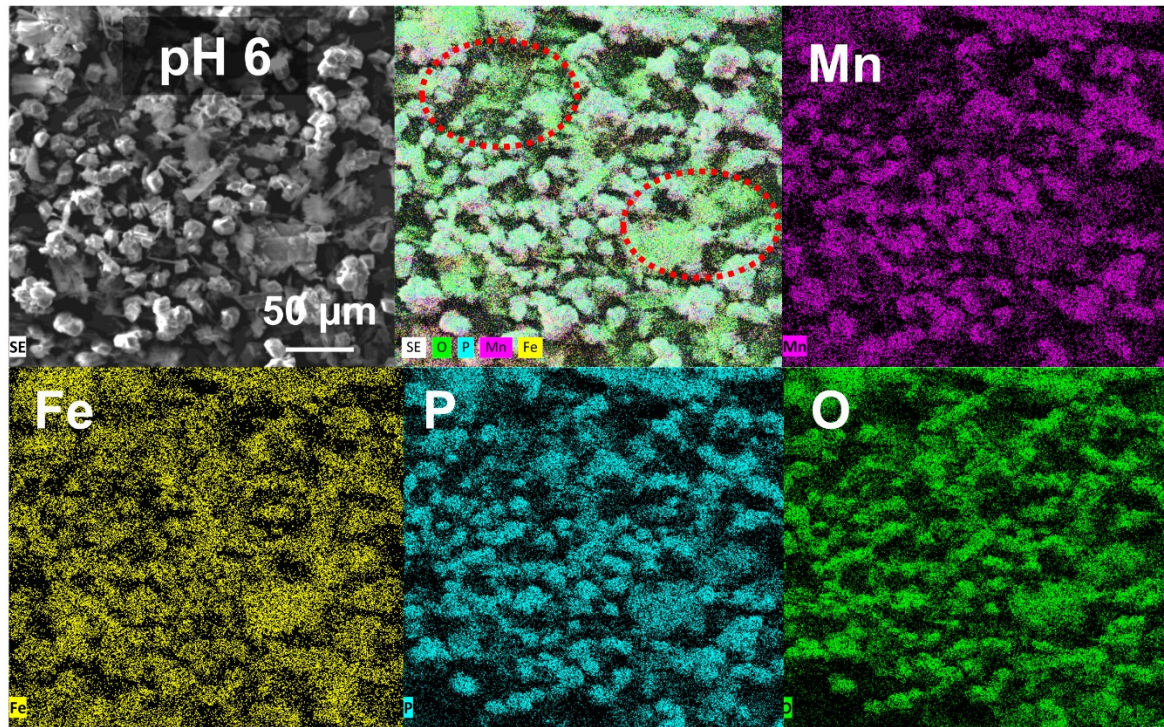


**Fig. S2** SEM-EDS images of the MFP precursors obtained at  $\text{pH} = 4$ , where the mapping shows the distribution of each element, including Mn, Fe, P, and O.



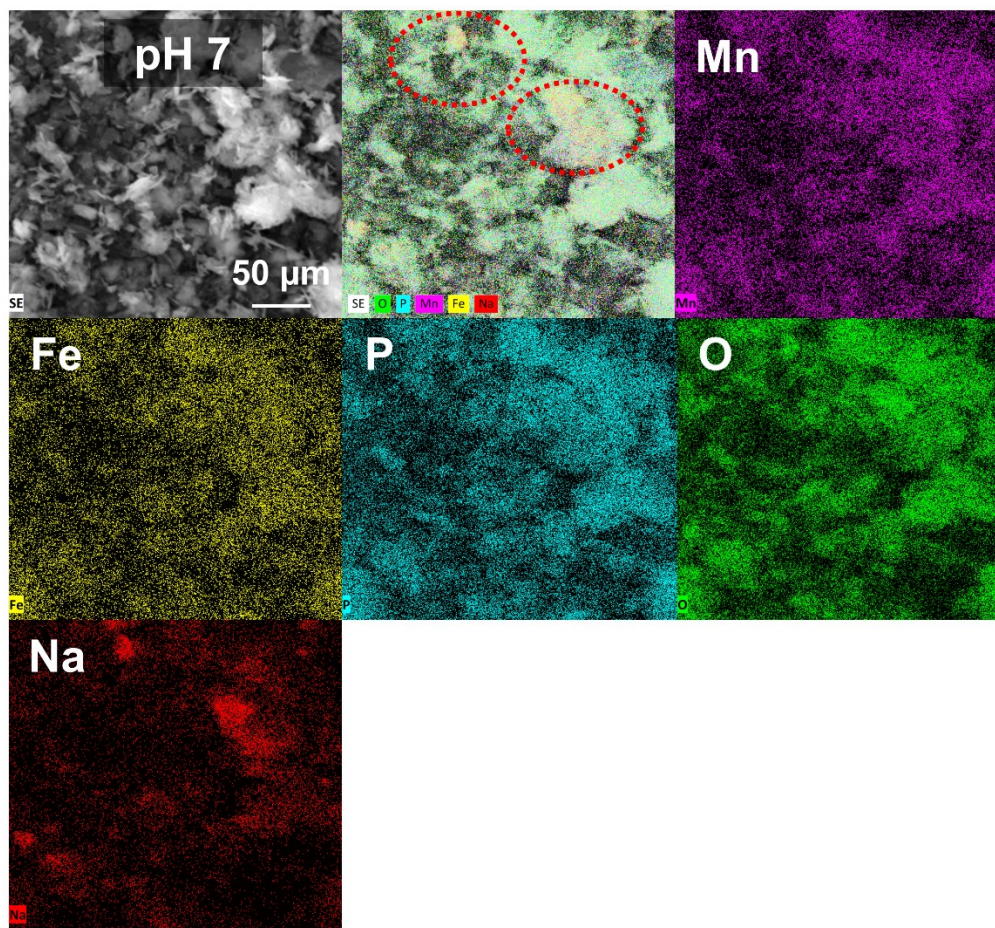


**Fig. S3** SEM-EDS images of the MFP precursors obtained at pH = 5, where the mapping shows the distribution of each element, including Mn, Fe, P, and O.

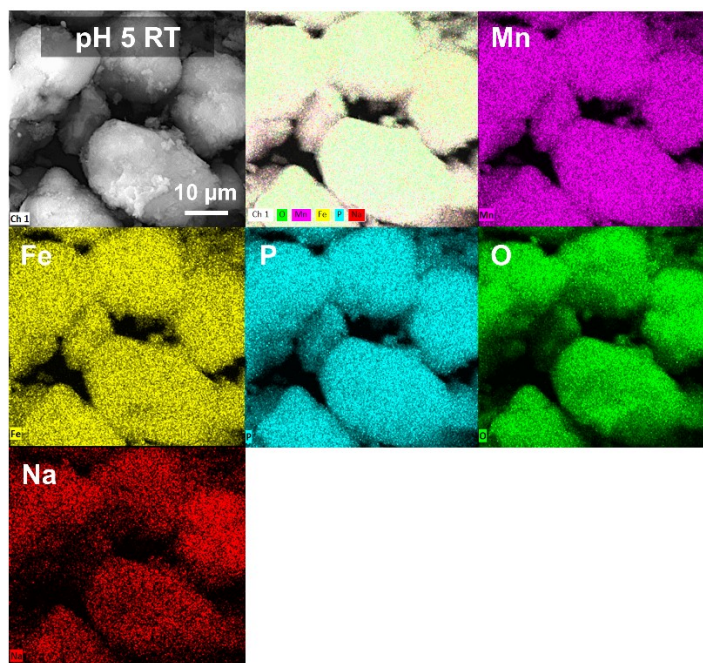


**Fig. S4** SEM-EDS images of the MFP precursors obtained at pH = 6, where the mapping shows the distribution of each element, including Mn, Fe, P, and O.

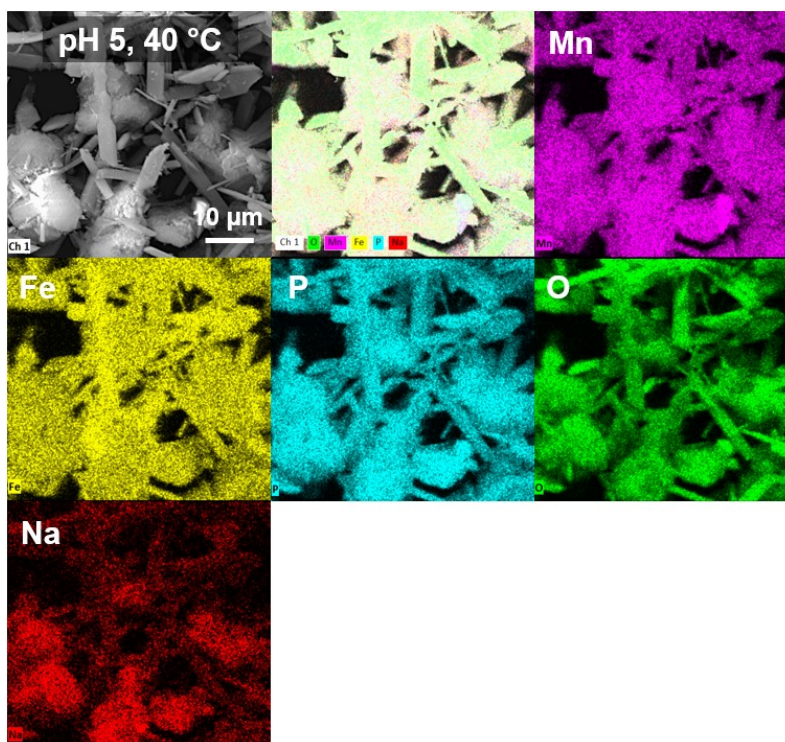




**Fig. S5** SEM-EDS images of the MFP precursors obtained at  $\text{pH} = 7$ , where the mapping shows the distribution of each element, including Mn, Fe, P, O, and Na.

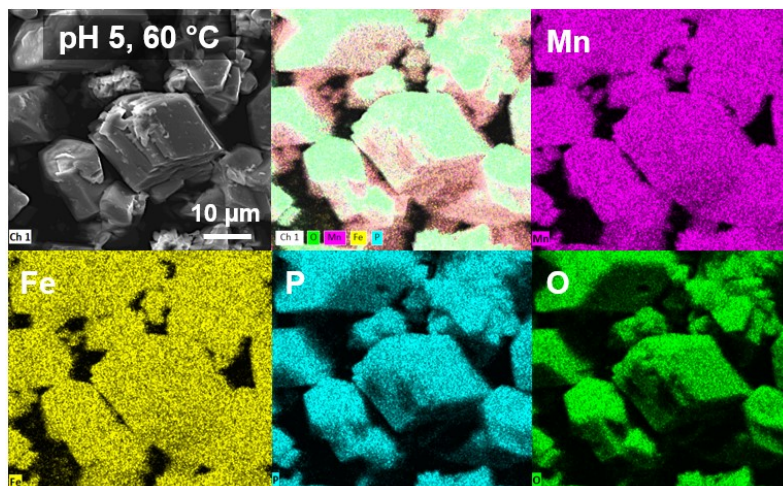


**Fig. S6** SEM-EDS images of the MFP precursors obtained at pH = 5 at room temperature (25 °C), where the mapping shows the distribution of each element, including Mn, Fe, P, O, and Na.

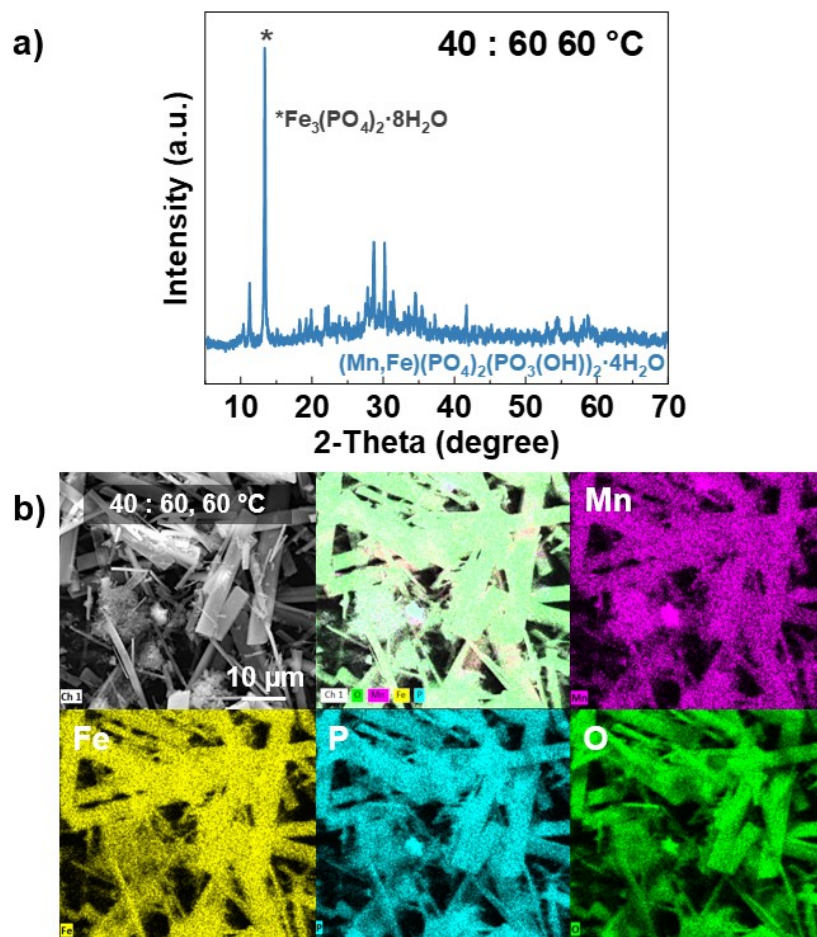


**Fig. S7** SEM-EDS images of the MFP precursors obtained at pH = 5 at 40 °C, where the mapping shows the distribution of each element, including Mn, Fe, P, O, and Na.

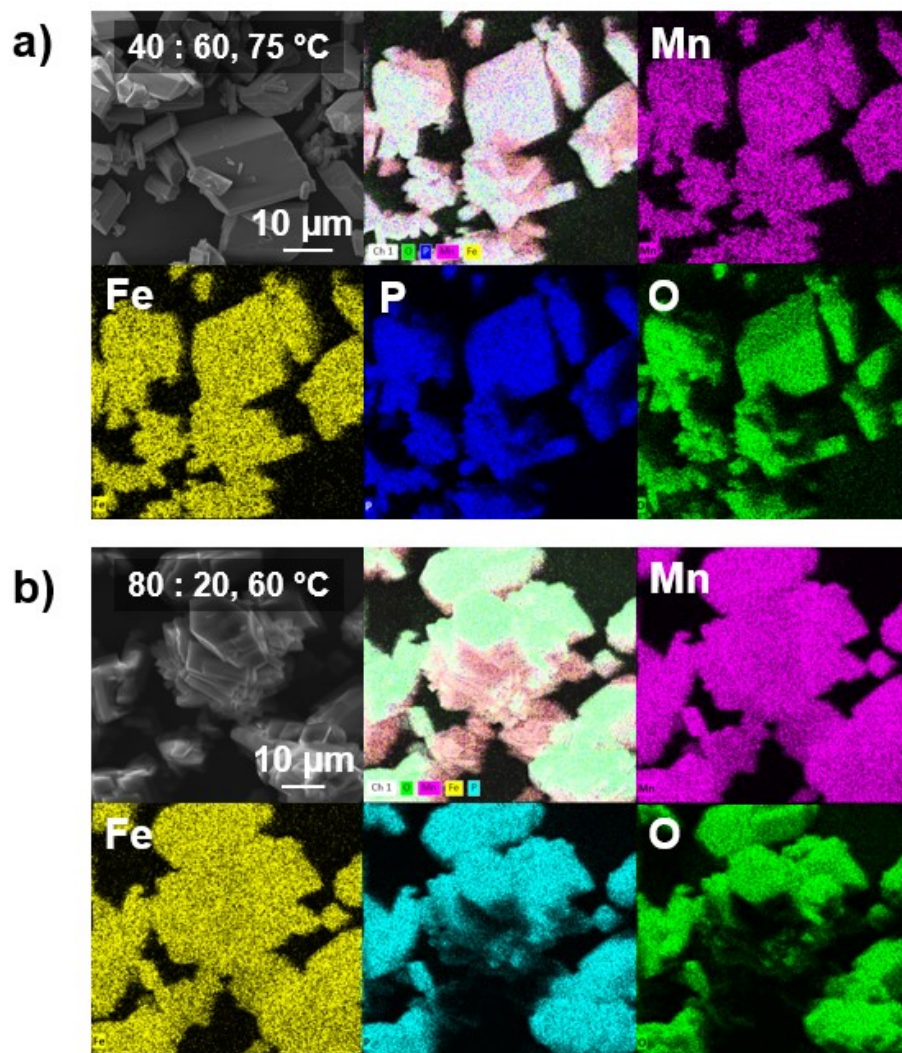




**Fig. S8** SEM-EDS images of the MFP precursors obtained at pH = 5 at 60 °C, where the mapping shows the distribution of each element, including Mn, Fe, P, and O.

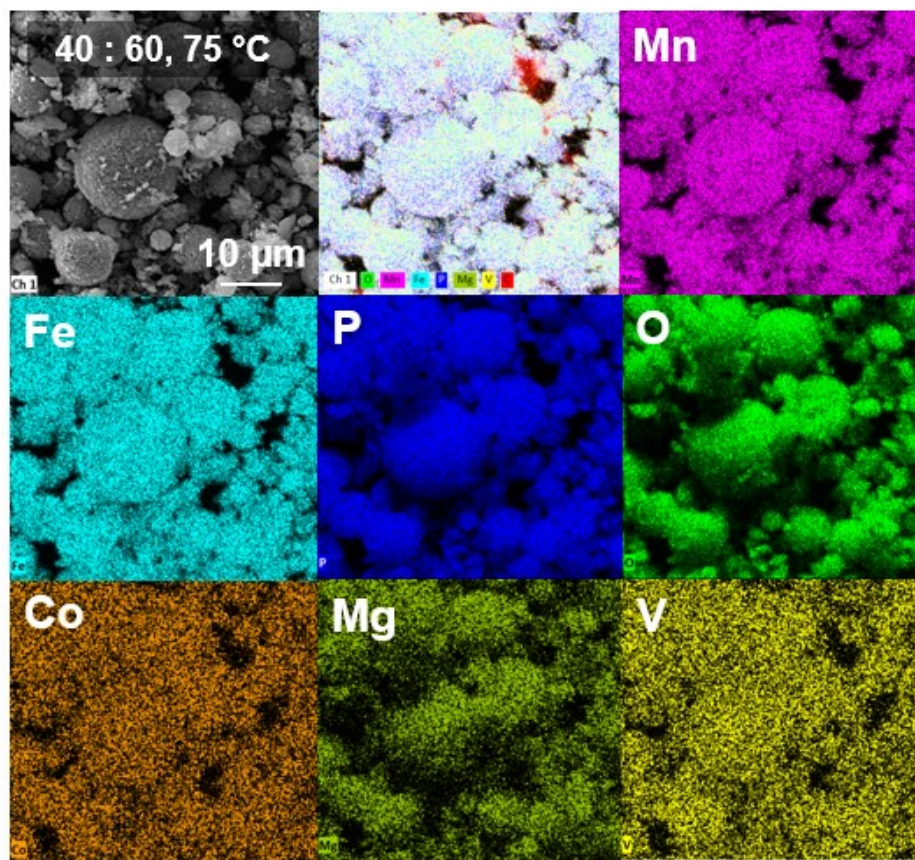


**Fig. S9** Morphological and structural analysis of the MFP precursor with an Mn : Fe ratio of 40 : 60 at 60 °C: (a) XRD pattern and (b) SEM-EDS mapping of each element, including Mn, Fe, P, and O.



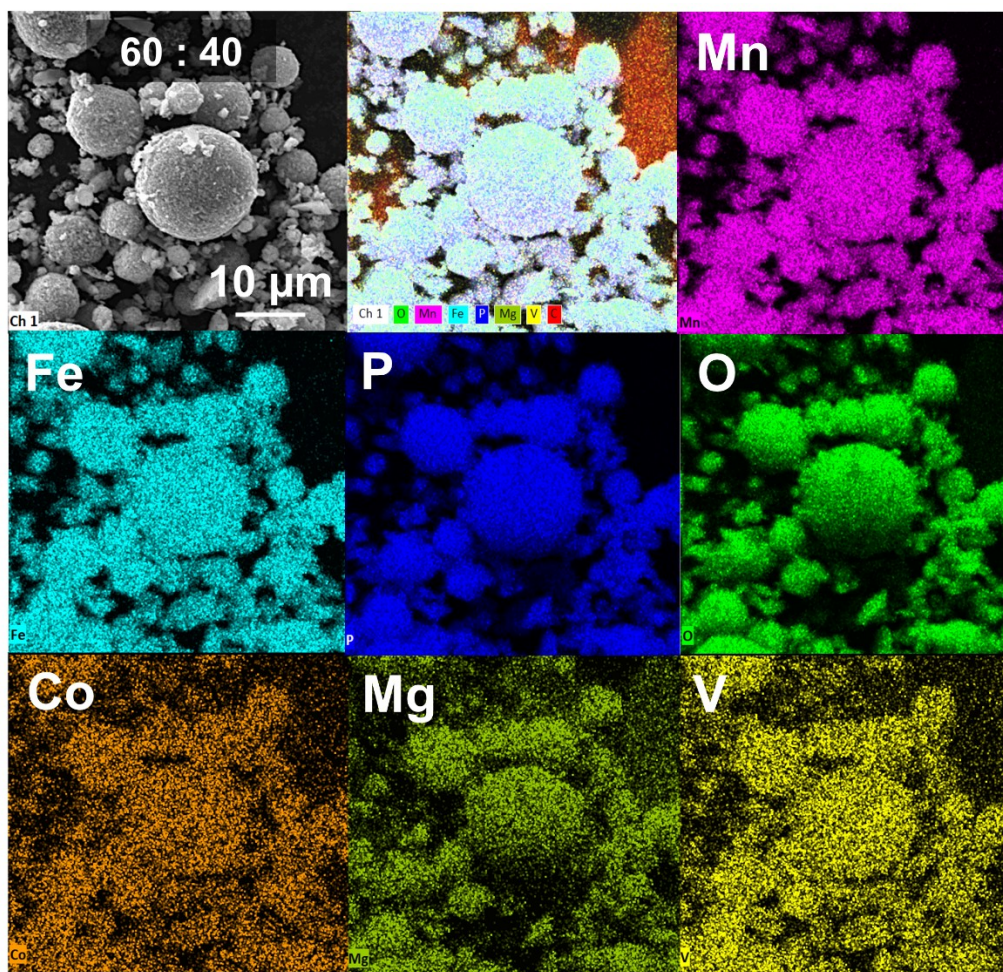
**Fig. S10** SEM-EDS mapping showing the distribution of each element, including Mn, Fe, P, and O for (a) Mn : Fe = 40 : 60 at 75 °C and (b) 80 : 20 at 60 °C.



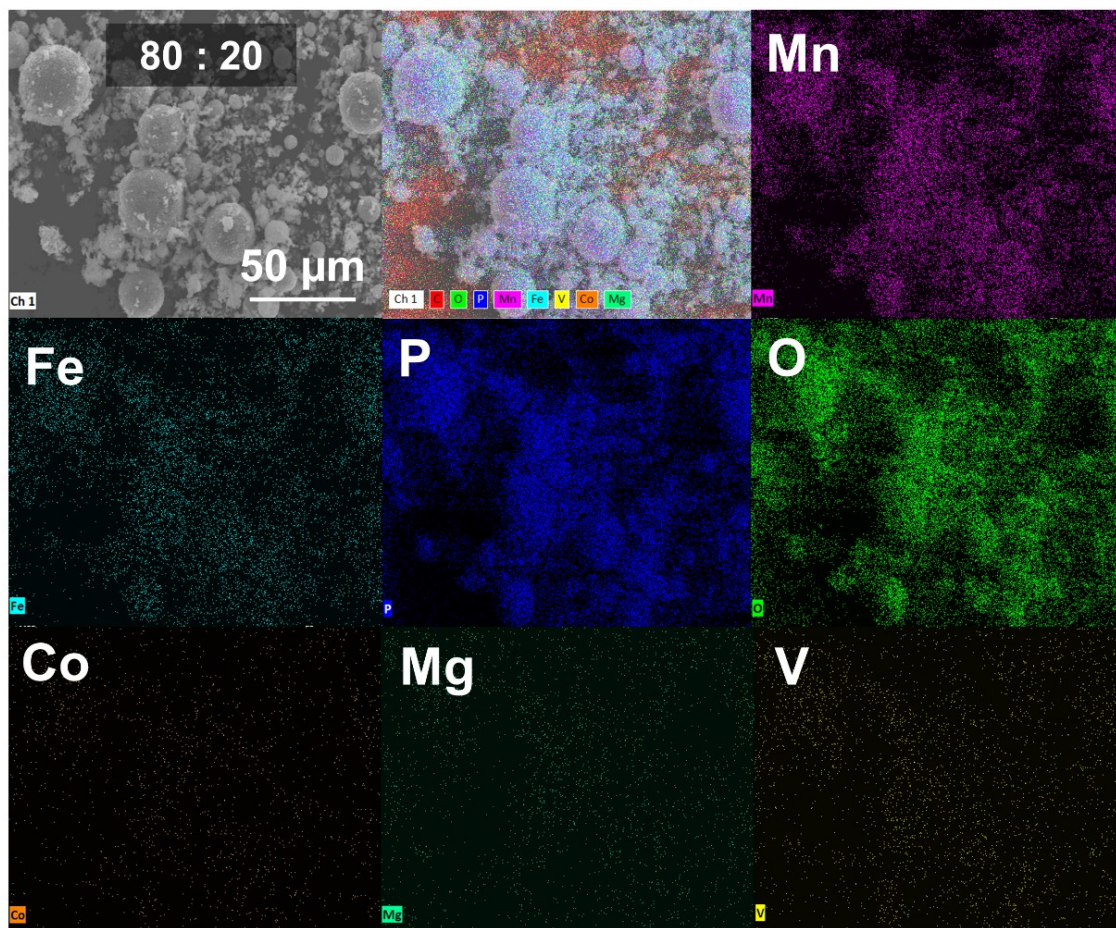


**Fig. S11** SEM-EDS mapping of the LMFP cathode with a Mn : Fe ratio of 40 : 60 at 75°C to examine the distribution of each element, including Mn, Fe, P, O, Co, Mg, and V.



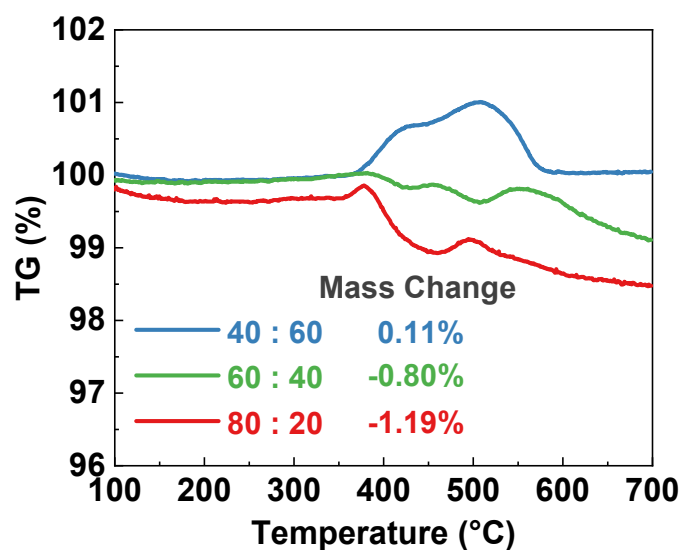


**Fig. S12** SEM-EDS mapping of the LMFP cathode with a Mn : Fe ratio of 60 : 40 to examine the distribution of each element, including Mn, Fe, P, O, Co, Mg, and V.

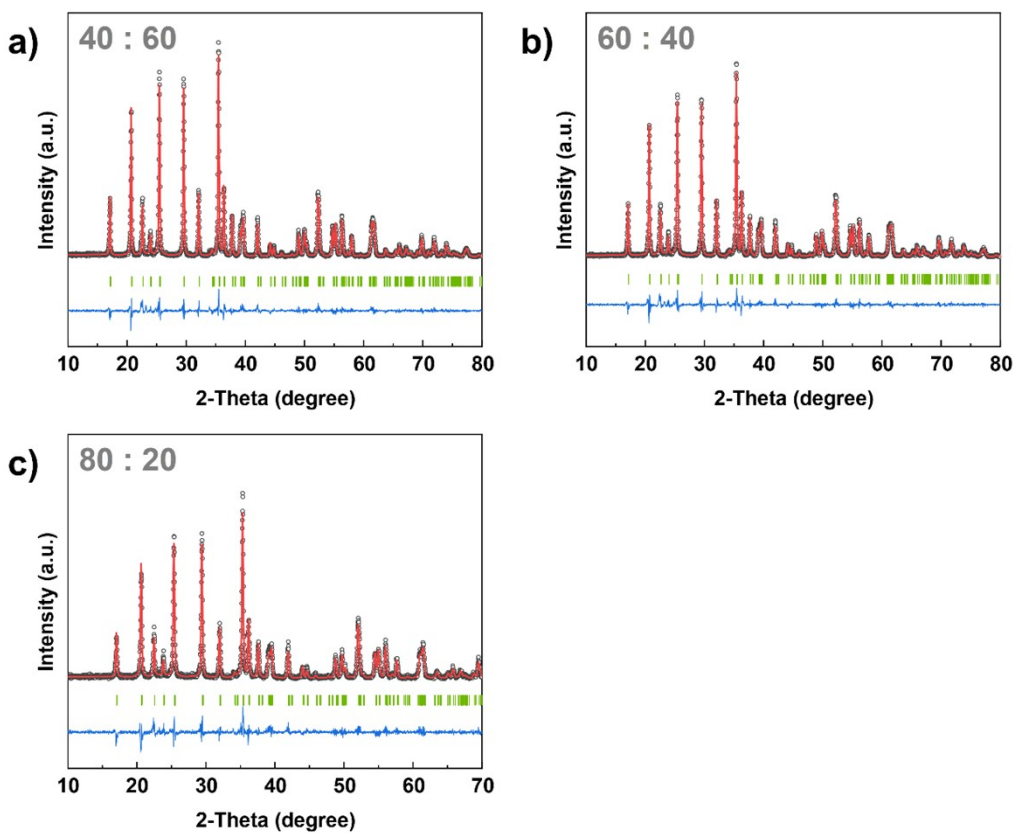


**Fig. S13** SEM-EDS mapping of the LMFP cathode with a Mn : Fe ratio of 80 : 20 to examine the distribution of each element, including Mn, Fe, P, O, Co, Mg, and V.

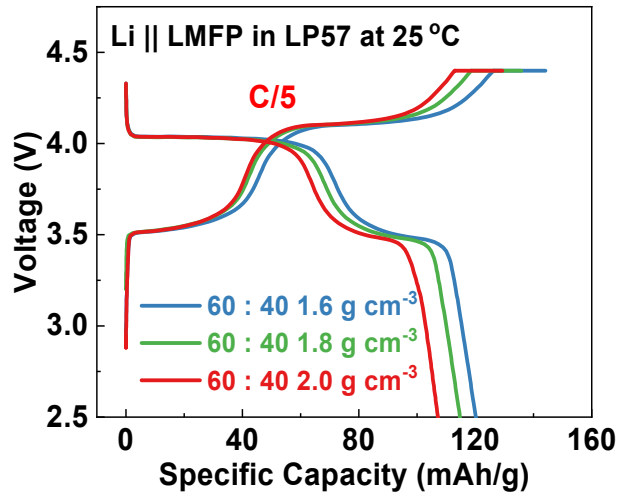




**Fig. S14** TGA profiles of the LMFP cathodes, showing the amount of carbon content for the 40 : 60, 60 : 40, and 80 : 20 samples



**Fig. S15** Rietveld refinement profiles of the LMFP cathodes with a Mn : Fe ratio of (a) 40 : 60, (b) 60 : 40, and (c) 80 : 20 with respect to the LMFP pattern (COD#2300354) as the reference pattern.



**Fig. S16** Charge-discharge profiles of the first cycle at C/5 rate for the Mn : Fe = 60 : 40 sample with different packing densities.



**Table S1** Elemental ratios of the precursors synthesized by the co-precipitation method as obtained from ICP-OES analysis with  $\text{FePO}_4$  as the reference

Sample	Mn	Fe	P
Target Ratio	0.600	0.400	0.67 for $\text{M}_2(\text{PO}_4)_3$ phase 0.80 for $(\text{Mn, Fe})_5(\text{PO}_4)_2(\text{HPO}_4)_2 \cdot 4\text{H}_2\text{O}$ phase 1.00 for $\text{NH}_4\text{MPO}_4$ phase
$\text{FePO}_4$ (Reference)	0.001	0.999	1.057
MFP- $\text{NH}_4\text{OH}$	0.608	0.392	1.030
MFP-KOH	0.593	0.407	0.805
MFP-NaOH	0.661	0.339	0.818
MFP-pH 4	0.679	0.321	0.850
MFP-pH 5	0.617	0.383	0.841
MFP-pH 5-CA	0.704	0.296	0.844
MFP-pH 7	0.606	0.394	0.711
MFP-pH 5-RT	0.588	0.412	0.746
MFP-pH 5-40 °C	0.524	0.476	0.722
MFP-pH 5-60 °C	0.620	0.380	0.828

**Table S2** Crystallographic data of LMFP (COD#2300354) used for performing the Rietveld refinement analysis

Parameter	$\text{LiMn}_{0.5}\text{Fe}_{0.5}\text{PO}_4$
Space Group	-P 2ac 2n (62)
a-axis (Å)	10.4072
b-axis (Å)	6.06188
c-axis (Å)	4.73357
$\alpha=\beta=\gamma$ (°)	90.00
V (Å <sup>3</sup> )	298.628
Radiation	Cu $K\alpha_1$ (1.5405 Å)

**Table S3** Structural information of LMFP (COD#2300354) used for performing the Rietveld refinement analysis

<b>Element</b>	<b>Label</b>	<b>x</b>	<b>y</b>	<b>z</b>	<b>B</b>	<b>Occupancy</b>
Li	Li1	0	0	0	0	0.996
Fe	Fe1	0.2828	0.25	0.9705	0	0.498
Mn	Mn1	0.2828	0.25	0.9705	0	0.498
P	P1	0.0952	0.25	0.4169	0	1
O	O1	0.0939	0.25	0.731	0	1
O	O2	0.4495	0.25	0.2062	0	1
O	O3	0.1604	0.0537	0.2778	0	1
Fe	Fe2	0	0	0	0	0.002
Mn	Mn2	0	0	0	0	0.002
Li	Li2	0.2828	0.25	0.9705	0	0.004