

## Supporting information

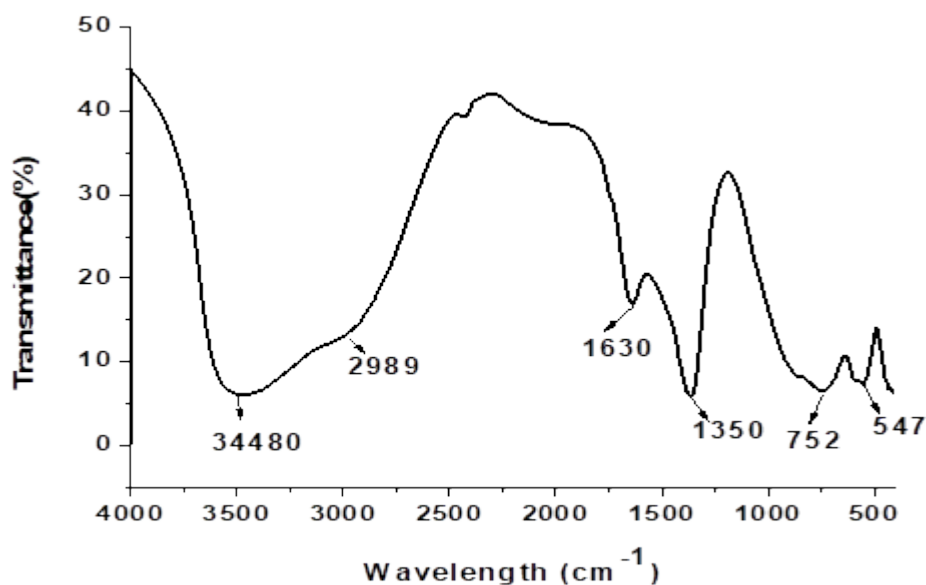


Fig.S<sub>1</sub> FTIR of CoAl-31

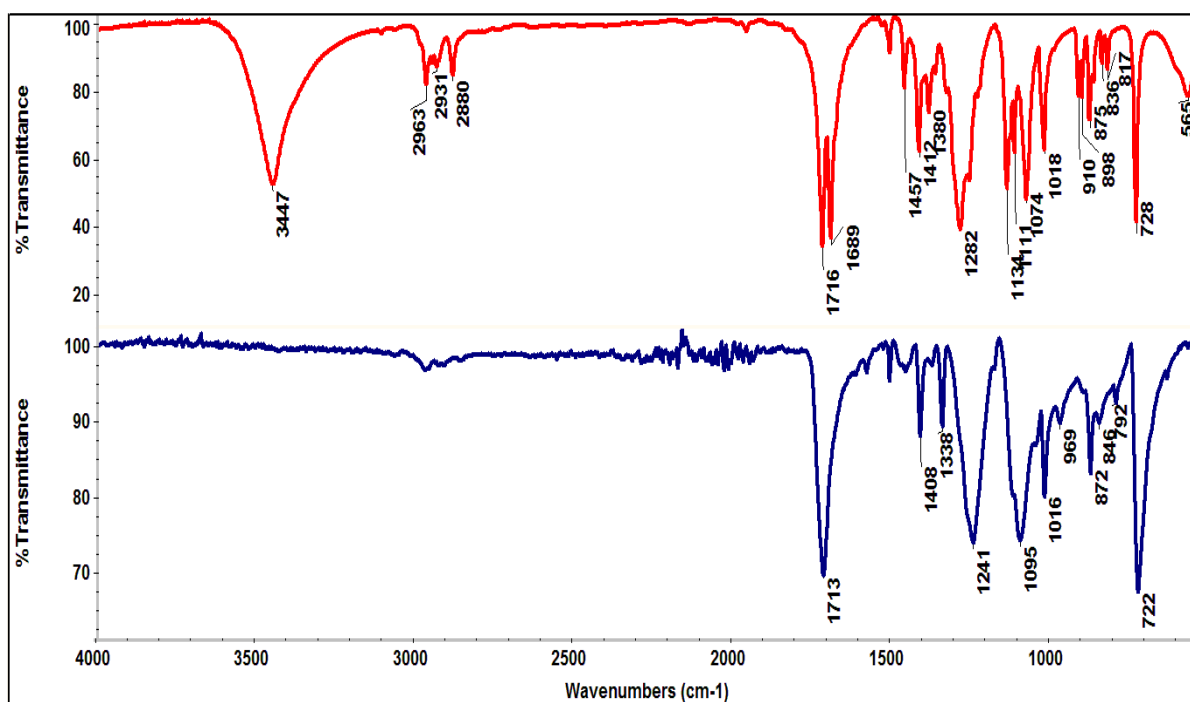
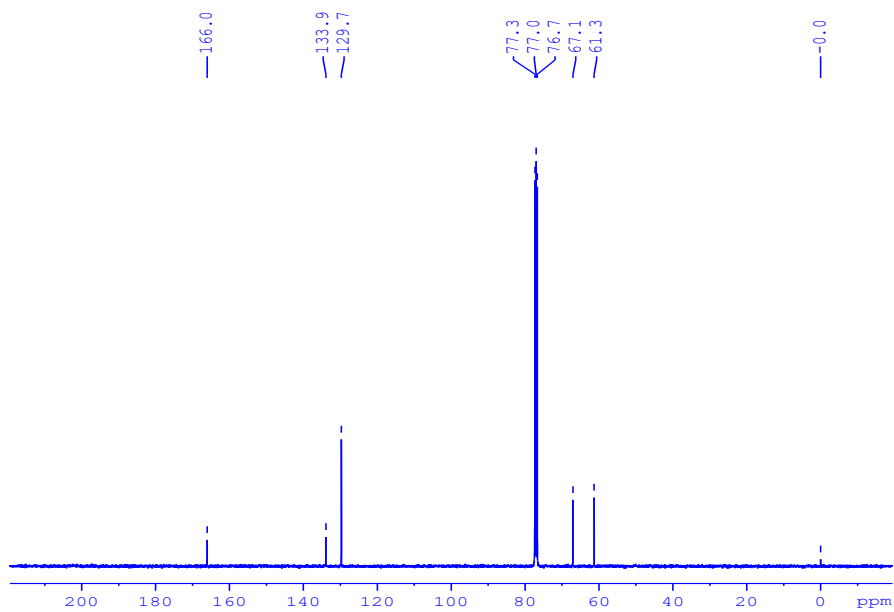
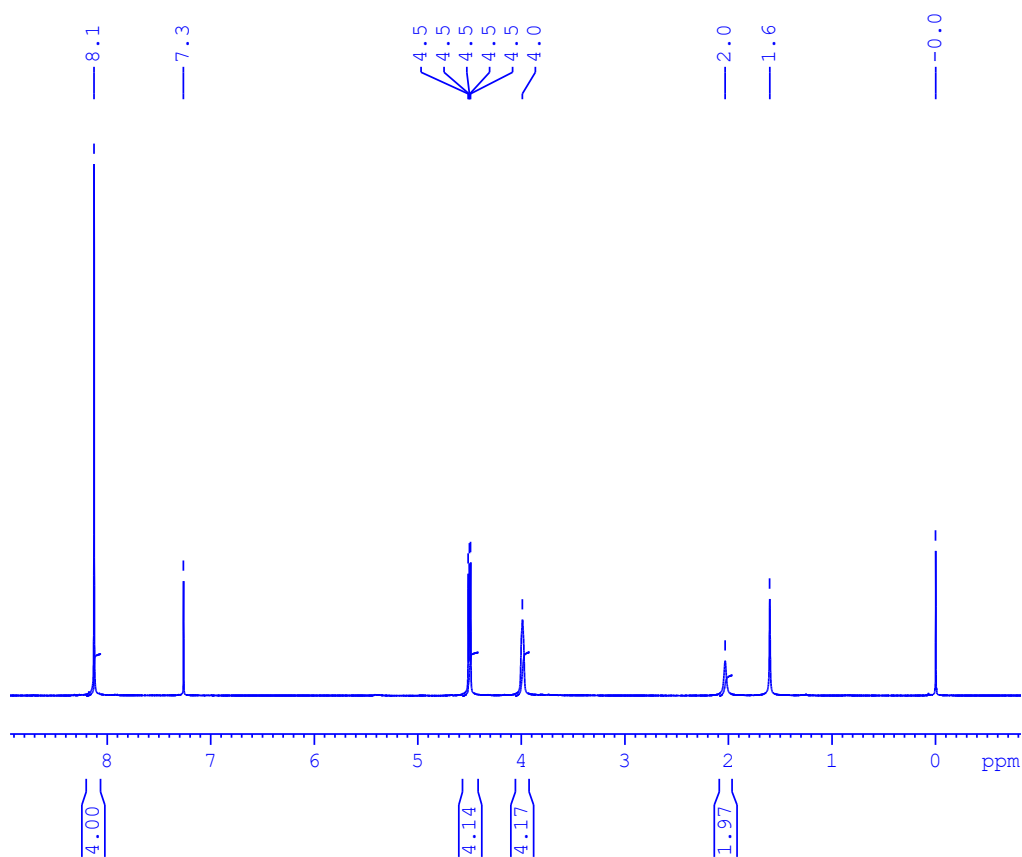


Fig.S<sub>2</sub> Overlaid FTIR spectrum of PET (bottom) and BHET (top)



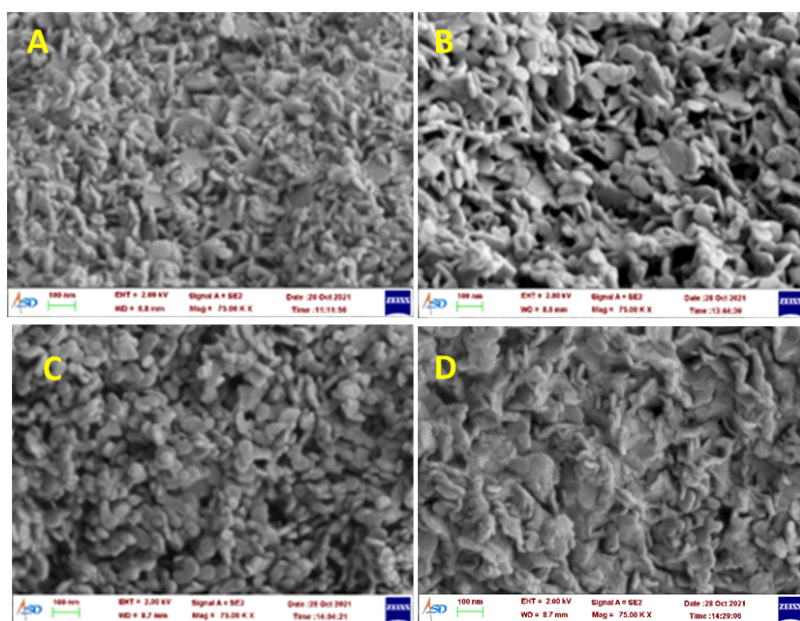
**Fig.S3  $^{13}\text{C}$  NMR of BHET**



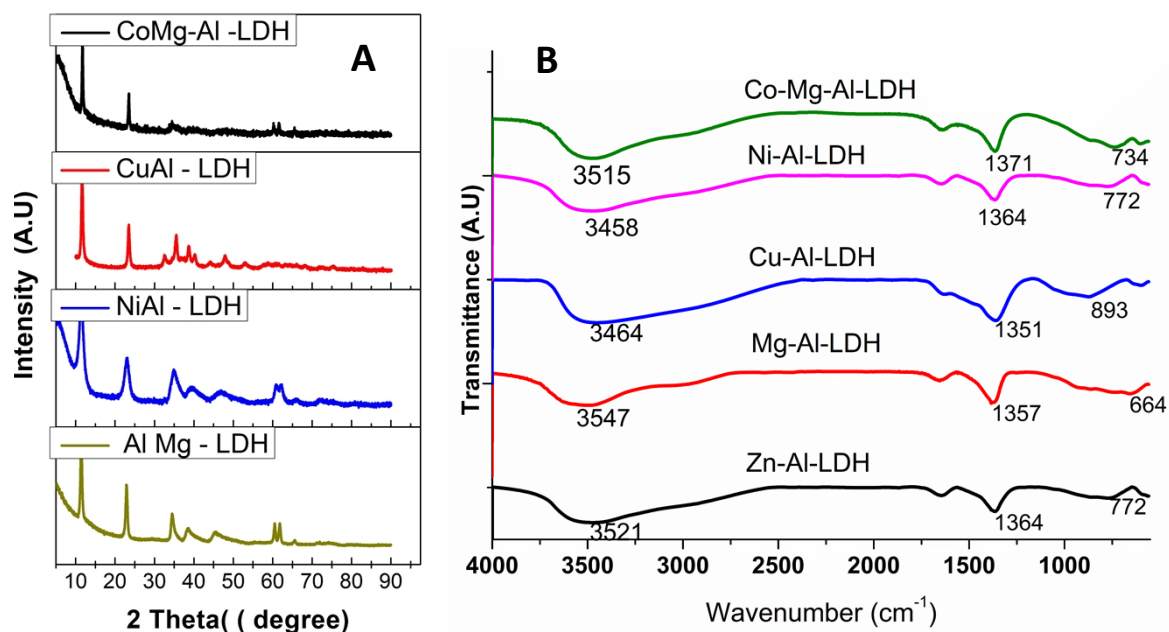
**Fig.S4  $^1\text{H}$  NMR of BHET**

**Table.S1 Properties of Co-Al CO<sub>3</sub> with different Co<sup>2+</sup>/Al<sup>3+</sup> ratios**

LDH	Experimental Co/Al Ratio	d003(Å)	d110(Å)	a(Å)	c(Å)	Crystalite size(nm)
CO-Al-11	0.8	7.50	1.52	3.04	22.50	13.0
CO-Al-21	1.9	7.53	1.53	3.06	22.59	13.0
CO-Al-31	2.7	7.66	1.54	3.08	22.98	10.4
CO-Al-41	3.6	7.69	1.55	3.10	23.07	9.8



**Fig.S5 FESEM images of Co-Al CO<sub>3</sub> with different Co<sup>2+</sup>/Al<sup>3+</sup> ratios (A) ratio 1:1, (B) ratio 2:1, (C) ratio 3:1 and (D) ratio 4:1**

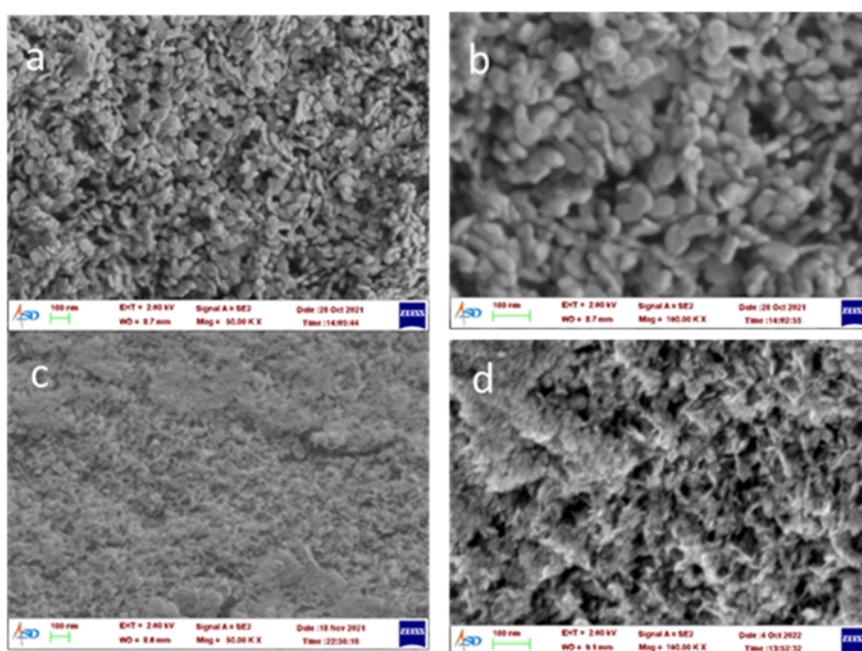


**Fig.S6 (A) XRD and (B) FTIR spectra of LDH materials with different M<sup>2+</sup> cation**

**Table S<sub>2</sub>: Comparison of Cobalt containing catalysts for PET glycolysis**

	Catalyst	Temperature (°C)	Time (h)	BHET (%)	Reference
1	Co <sub>3</sub> O <sub>4</sub>	260	1	63	<a href="https://doi.org/10.1016/j.polydegradstab.2013.01.007">https://doi.org/10.1016/j.polydegradstab.2013.01.007</a>
2	CoCl <sub>2</sub> (anh)	190	3	65	<a href="https://doi.org/10.1007/s10562-016-1897-0">https://doi.org/10.1007/s10562-016-1897-0</a> .
	CoCl <sub>2</sub> (anh)/dcype			71	
	CoCl <sub>2</sub> (anh)/dppe			53	
	CoCl <sub>2</sub> (anh)/dppf			31	
	CoCl <sub>2</sub> (anh)/BEt <sub>3</sub>			85	
3	Recovered CoO from lithium-ion batteries	196	2	10	<a href="https://doi.org/10.1007/s12649-019-00807-6">https://doi.org/10.1007/s12649-019-00807-6</a>
4	Co NPs	180	3	77	<a href="https://doi.org/10.1021/acssuschemeng.8b02294">https://doi.org/10.1021/acssuschemeng.8b02294</a>
5	CoZnO	196	2	80	<a href="https://doi.org/10.1007/s12649-019-00807-6">https://doi.org/10.1007/s12649-019-00807-6</a>
6	rGO/[TESPMI] <sub>2</sub> CoCl <sub>4</sub>	190	3	95	<a href="https://doi.org/10.1016/j.polydegradstab.2021.109691">https://doi.org/10.1016/j.polydegradstab.2021.109691</a>

7	CoMn <sub>2</sub> O <sub>4</sub>	260	1	89	<a href="https://doi.org/10.1016/j.polymdegradstab.2013.01.007">https://doi.org/10.1016/j.polymdegradstab.2013.01.007</a>
8	ZnCo <sub>2</sub> O <sub>4</sub>	260	1	89	<a href="https://doi.org/10.1016/j.polymdegradstab.2013.01.007">https://doi.org/10.1016/j.polymdegradstab.2013.01.007</a>
9	CoFe <sub>2</sub> O <sub>4</sub> @ZIF-8/ZIF-67	200	1	84	<a href="https://doi.org/10.1016/j.fuel.2021.121397">https://doi.org/10.1016/j.fuel.2021.121397</a>
10	CoFe <sub>2</sub> O <sub>4</sub>	195	2.5	73	<a href="https://doi.org/10.1016/j.eurpolymj.2021.110590">https://doi.org/10.1016/j.eurpolymj.2021.110590</a>
11	CoFe <sub>2</sub> O <sub>4</sub> /C10-OAC	195	2.5	96	<a href="https://doi.org/10.1016/j.eurpolymj.2021.110590">https://doi.org/10.1016/j.eurpolymj.2021.110590</a>
12	Mechano chemically prepared CoFe <sub>2</sub> O <sub>4</sub>	190	6.0	77	<a href="https://doi.org/10.1016/j.cej.2022.137926">https://doi.org/10.1016/j.cej.2022.137926</a>
13	Co Al 31	180	2	96	This work
13	Co Al 31@Fe <sub>3</sub> O <sub>4</sub>	180	2	99	This work



**Fig.S7 FESEM images of (a, b) CoAl-31 LDH and (c, d) NiAl-31 LDH**

**Table S<sub>3</sub> Comparison of Magnetically separable catalysts for PET glycolysis**

	Catalyst	BHET yield (%)	Temperature	Time (H)	Reference
1	Superparamagneti	90	300, 1.1 MPa	1	<a href="https://doi.org/10.1039/C3G">https://doi.org/10.1039/C3G</a>

	c $\gamma$ -Fe <sub>2</sub> O <sub>3</sub> Nanoparticles				C41834K
2	$\gamma$ -Fe <sub>2</sub> O <sub>3</sub> /nitrogen-doped graphene hybrid material	100	195	3	<a href="http://dx.doi.org/10.1016/j.polyimdegadstab.2017.08.033">http://dx.doi.org/10.1016/j.polyimdegadstab.2017.08.033</a>
3	Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> @(mim)[FeCl <sub>4</sub> ]	63	170	24	<a href="https://doi.org/10.1016/j.apcatb.2019.118110">https://doi.org/10.1016/j.apcatb.2019.118110</a>
4	$\gamma$ -Fe <sub>2</sub> O <sub>3</sub>	40	195	3	<a href="http://dx.doi.org/10.1016/j.polyimdegadstab.2017.08.033">http://dx.doi.org/10.1016/j.polyimdegadstab.2017.08.033</a>
5	h-BNNS with Fe <sub>3</sub> O <sub>4</sub> nanoparticles	100	200, in autoclave		<a href="https://doi.org/10.1016/j.polyimdegadstab.2019.108962">https://doi.org/10.1016/j.polyimdegadstab.2019.108962</a>
6	Fe <sub>3</sub> O <sub>4</sub> -boosted MWCNT	100	190	2	<a href="https://doi.org/10.1039/C6GC00534A">https://doi.org/10.1039/C6GC00534A</a> .
7	ZnO-Fe <sub>3</sub> O <sub>4</sub> magnetic hollow micro-sized nanoaggregates	92.3	190	0.5	<a href="https://doi.org/10.1039/D3GC01762A">https://doi.org/10.1039/D3GC01762A</a>
8	Mg-Al-O@Fe <sub>3</sub> O <sub>4</sub>	31.4	240	1.5	<a href="https://doi.org/10.1016/j.wasman.2021.03.049">https://doi.org/10.1016/j.wasman.2021.03.049</a>
9	two-dimensional Fe <sup>III</sup> nanosheets	100	200	0.5	<a href="https://doi.org/10.1039/D0RE00385A">https://doi.org/10.1039/D0RE00385A</a>
10	Fe <sub>3</sub> O <sub>4</sub> nanodispersions	93	210	0.5	<a href="https://doi.org/10.1021/acscuschemeng.3c01206">https://doi.org/10.1021/acscuschemeng.3c01206</a>
11	Fe <sub>3</sub> O <sub>4</sub> -CP	93.5	190	2	<a href="https://doi.org/10.1039/D3GC01707A">https://doi.org/10.1039/D3GC01707A</a>
12	Coal @Fe <sub>3</sub> O <sub>4</sub>	99	180	2	This work

**Table S<sub>4</sub> ICP-AES analysis of Co-Al 31@Fe<sub>3</sub>O<sub>4</sub>**

<b>Element</b>	<b>Wt.% (from ICP-AES)</b>
<b>Cobalt</b>	29.8
<b>Aluminum</b>	4.7
<b>iron</b>	18.2