Supporting Information

Alkali-Free Hydrothermally Reconstructed NiAl Layered Double Hydroxides for Catalytic Transesterification

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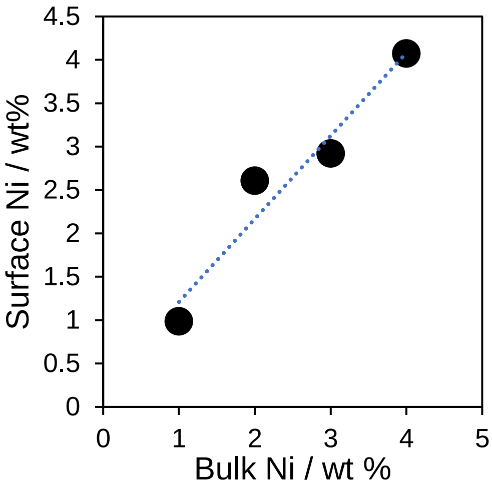
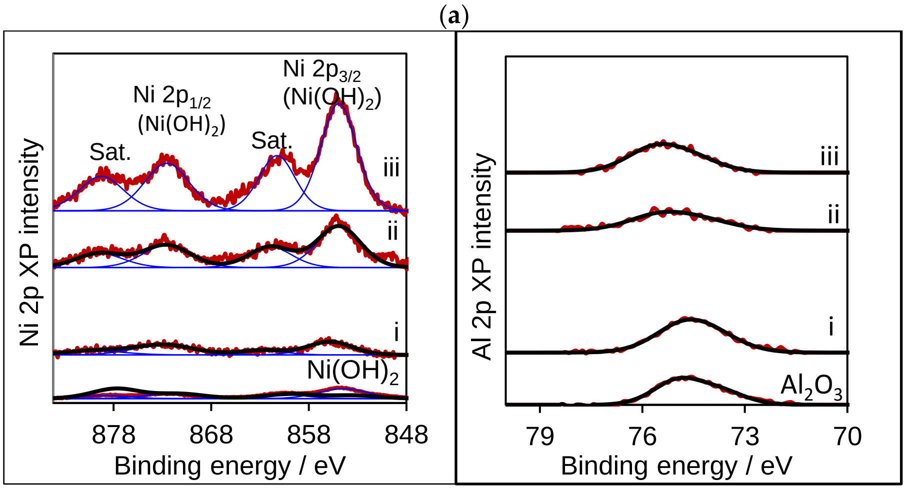
1. Characterisation



(a)



(b)

Figure S1. (**a**) Ni surface and bulk loading of NiAl LDHs determined by XPS and EDS respectively; (**b**) High-resolution XP spectra of NiAl LDHs with nominal Ni:Al atomic ratios of (i) 1.5, (ii) 3 and (iii) 4.

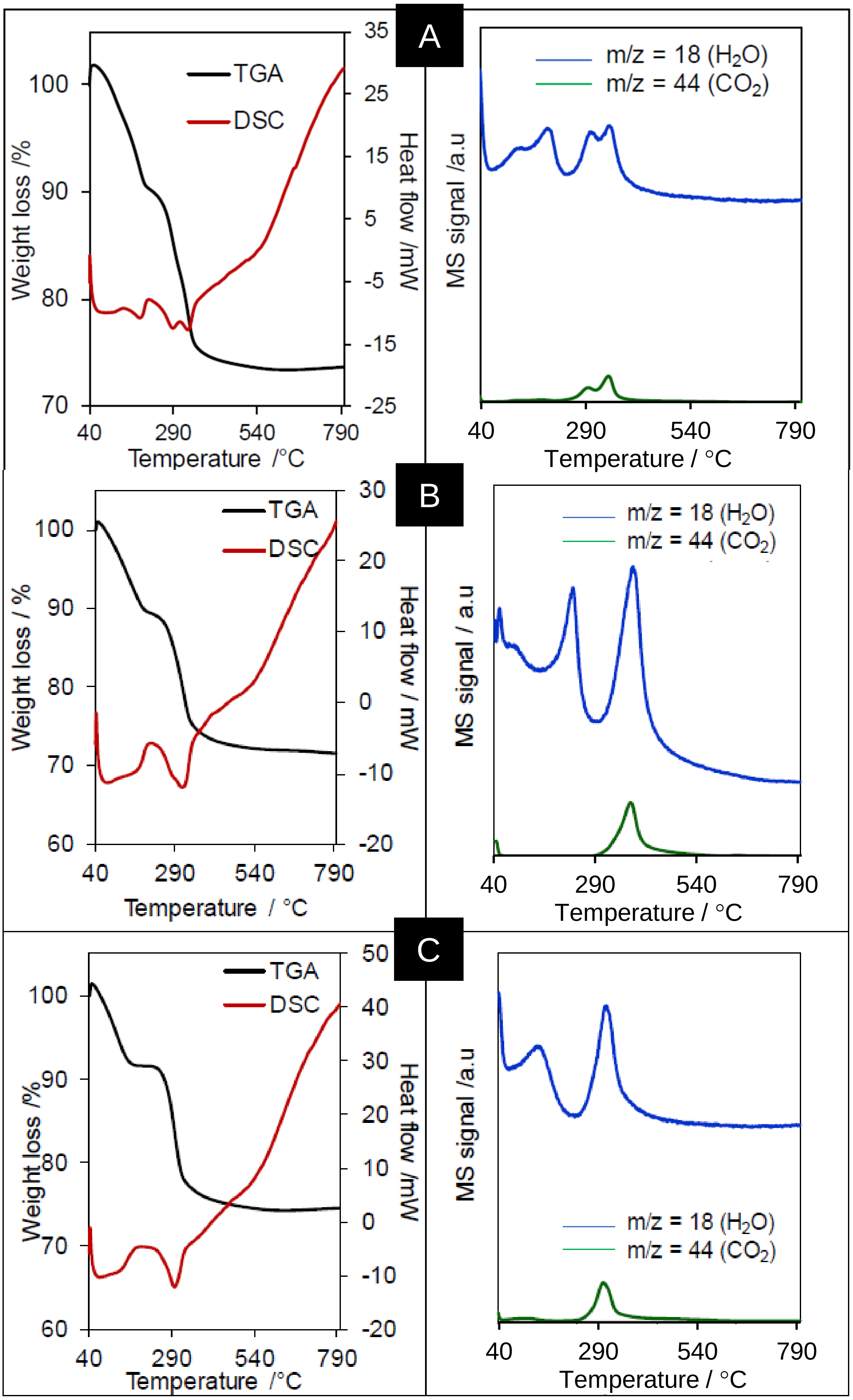


Figure S2. Thermogravimetric analysis of NiAl LDHs with nominal Ni:Al atomic ratios of (**A**) 1.5, (**B**) 3 and (**C**) 4.

Table S1. Weight losses from TGA.

Ni:Al ratio[a]	Weight loss / %		Total weight loss / %
	H_2O	CO_3^{2-}	
1.7:1	11.0	9.1	20.1
2.7:1	11.3	12.5	23.6
4.1:1	14	9	24.1

[a]EDS.

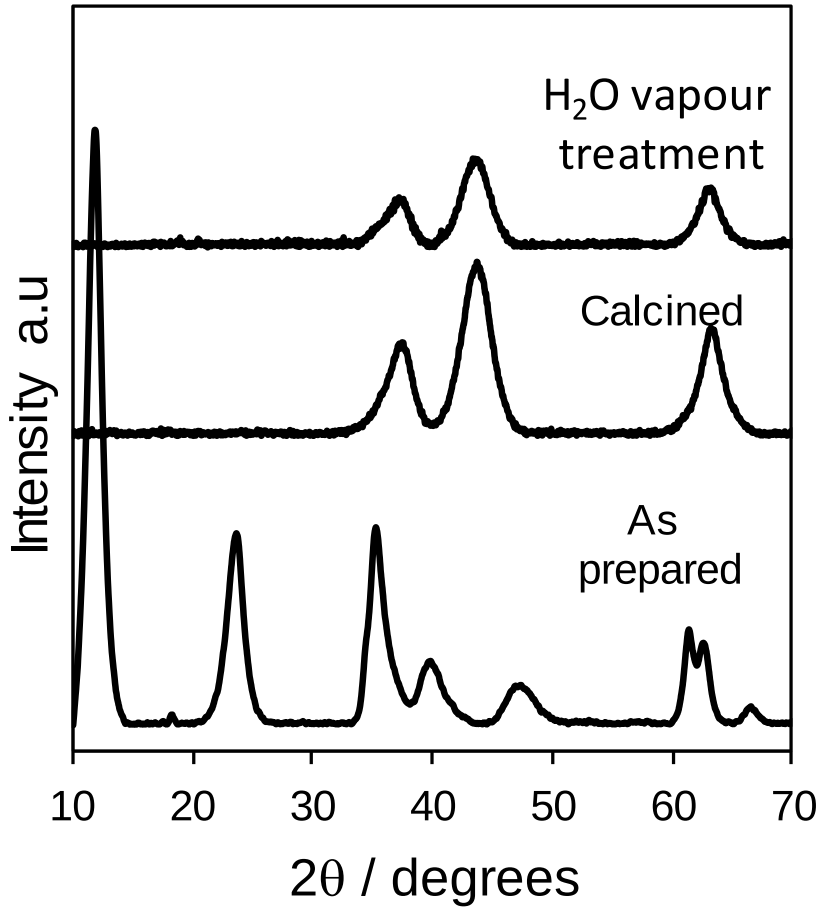


Figure S3. Powder XRD patterns of Ni4Al LDH after different hydrothermal treatments.

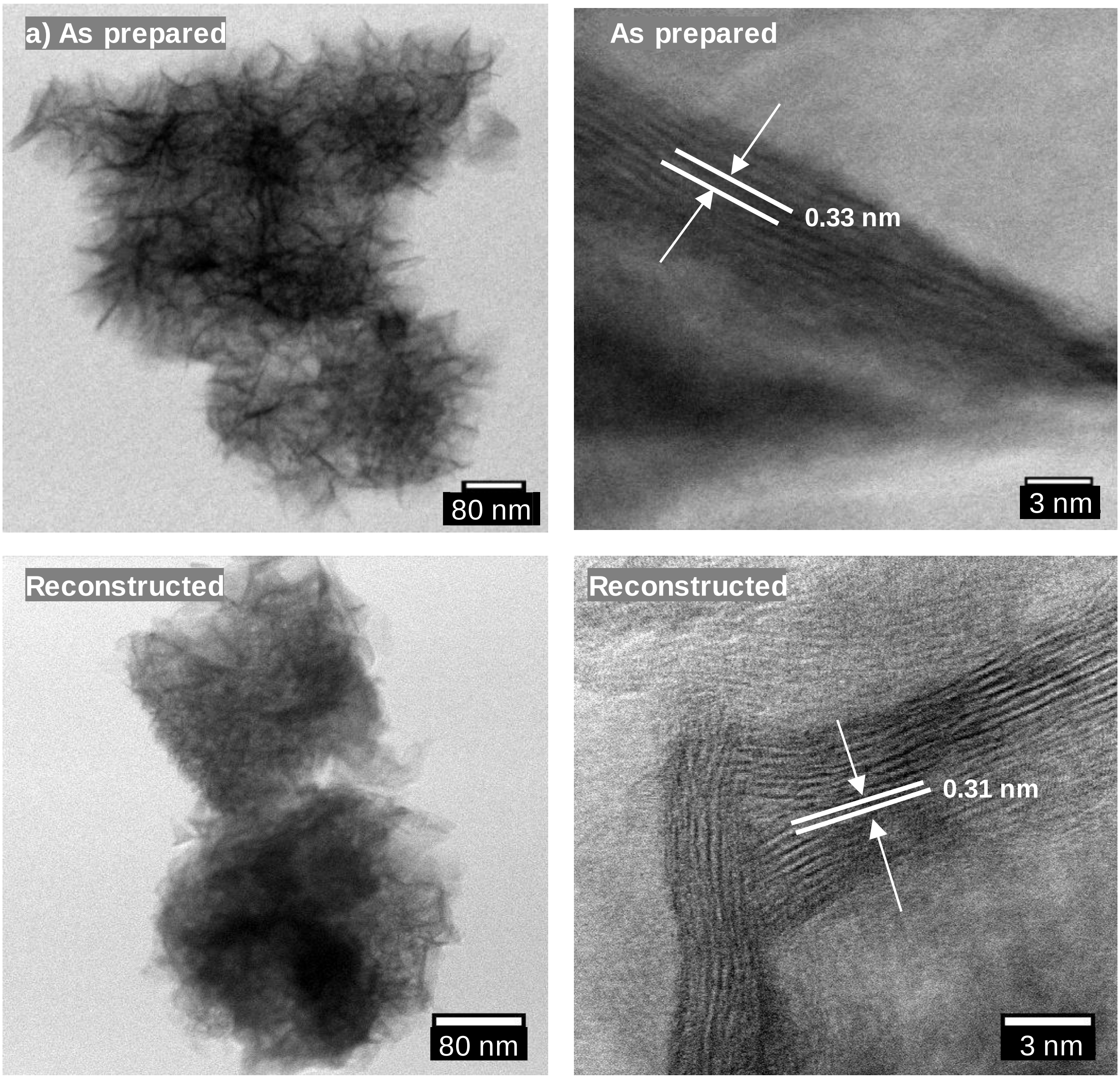


Figure S4. HRTEM images of (top) precipitated and (bottom) hydrothermally reconstructed $Ni_{4.1}Al$ LDH. Hydrothermal treatment involved 350 °C calcination and subsequent 110 °C water treatment.

2. Transesterification reactions

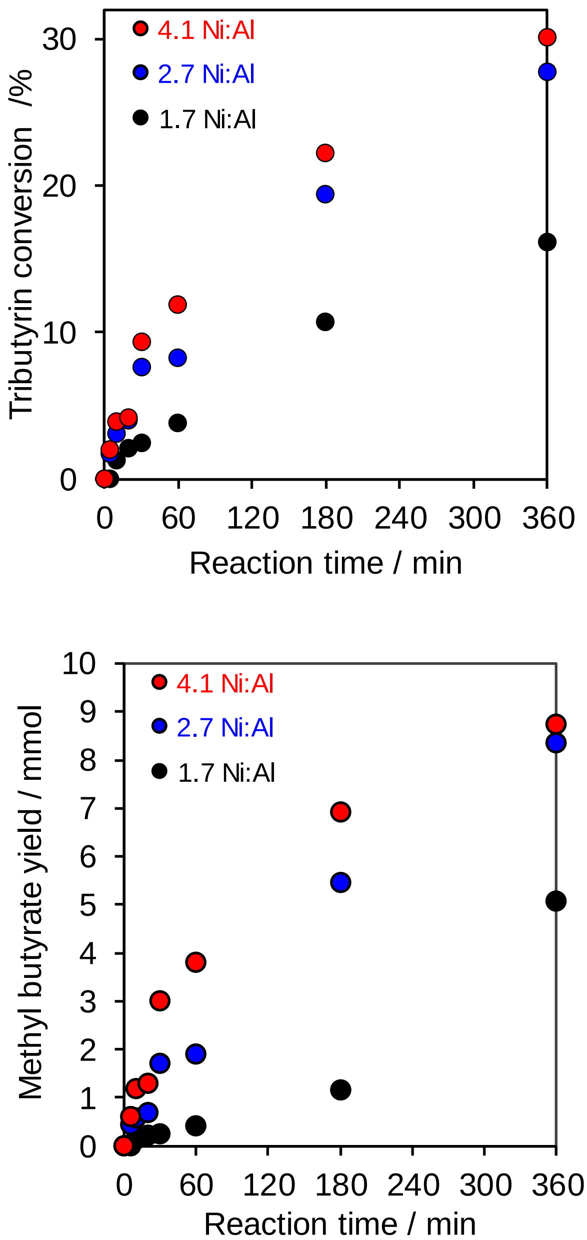


Figure S5. Reaction profiles for C4 TAG transesterification with methanol by NiAl LDH as a function of Ni:Al atomic ratio. Reaction conditions: 100 mg catalyst, 10 mmol of triglyceride (TAG), 308 mmol (12.5 ml) methanol with 0.0025 mol (0.59 cm^3) dihexylether as internal standard. Reaction was done at 650 rpm, 110 °C for 24 hours.

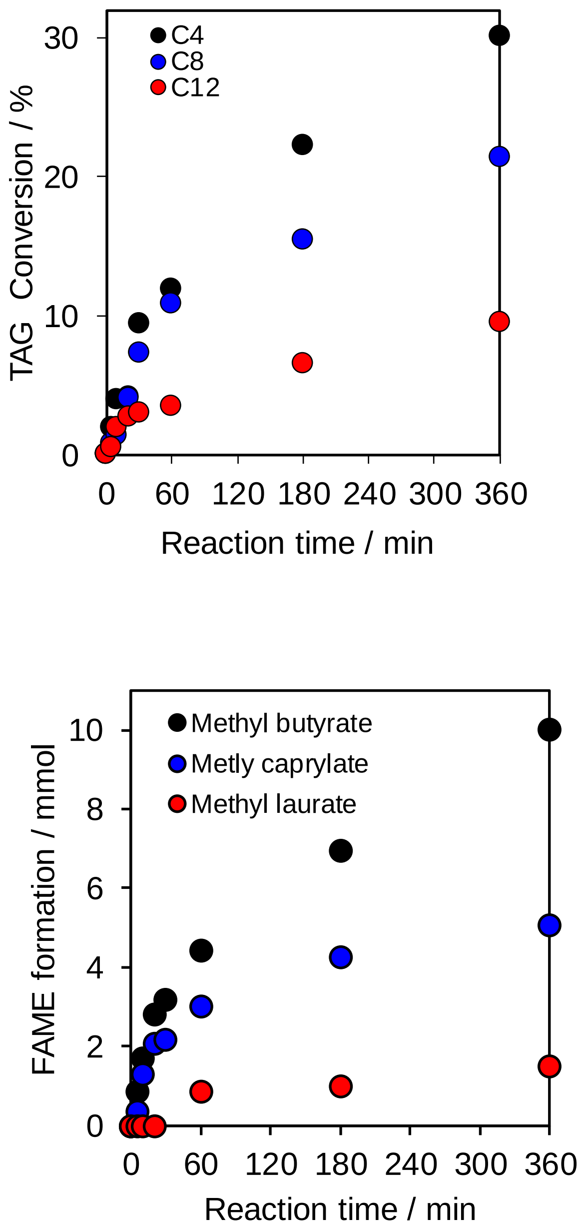


Figure S6. Reaction profiles for C_4-C_{12} TAG transesterification with methanol by reconstructed $Ni_{4.1}Al$ LDH. Reaction conditions: 100 mg catalyst, 10 mmol of triglyceride (TAG), 308 mmol (12.5 ml) methanol with 0.0025 mol (0.59 cm^3) dihexylether as internal standard. 20 wt% 1-butanol was introduced for longer chain C8-C12 TAGs (tricaprylin and trilaurin) to improve their miscibility. Reaction was performed at 110 °C, 650 rpm, for 24 hours.

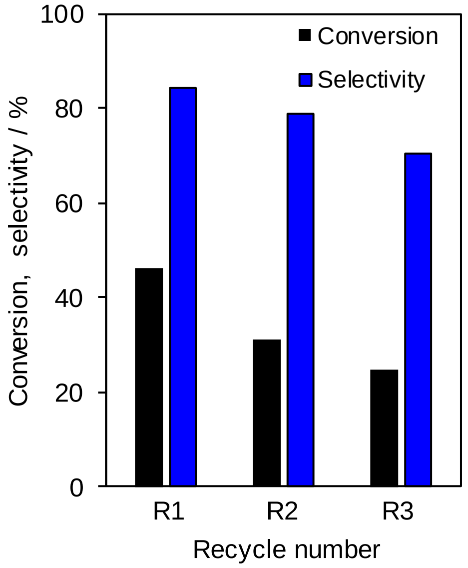


Figure S7. Recycling of hydrothermally reconstructed Ni4Al LDH in the transesterification of tributyrin with methanol. Reactions conditions: 100 mg catalyst, 110 °C, 10 mmol tributyrin (TAG), 30:1 methanol:TAG molar ratio, 650 rpm stirring, 24 h reaction.

Table S2. ICP-OES analysis on reconstructed Ni_4Al LDH after 3 recycles.

Sample	**Atomic concentration / %**		**Ni:Al**
	Ni	**Al**	
Fresh catalyst	64.96	15.80	4.11
Catalyst after 3rd cycle	55.84	13.71	4.07