**Supporting information** 

## Selective conversion of lactic acid to acrylic acid over alkali and alkaline-earth metal co-modified NaY zeolites

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Fig. S1 XRD patterns of various modified NaY catalysts.



Fig. S2 (A) Catalytic performance as a function of time-on-stream during LA conversion and (B) XRD of the various catalysts calcined at different temperatures.



Fig. S3 (A) <sup>29</sup>Si and (B) <sup>27</sup>Al MAS NMR spectra of various modified catalysts.

<sup>27</sup>Al MAS NMR spectra of several NaY-based catalysts are shown in Fig. S2. A broad peak at 59 ppm on the parent NaY is attributed to the tetrahedral coordinated aluminum in the zeolite framework, which is charge balanced by sodium ion. This peak becomes narrow after etching, and the position of the peak slightly shifts to 61 ppm. No significant changes in <sup>27</sup>Al MAS NMR spectrum for the fresh and regenerated catalyst, indicating negligible change of the framework Al environment.



Fig. S4 Fitting of the experimental Ca K-edge XANES data of (A) KOH-Ca-NaY regen 4x and (B) KOH-Ca-ENaY regen 1x.



Fig. S5 FTIR spectra of Spent, Fresh and Regenerated KOH-Ca-NaY catalysts.



Fig. S6 Thermal gravimetric analysis of the spent (after 77h TOS) and regenerated catalysts.

Samples	<sup>29</sup> Si NMR					ICP				
	Si/Al	n=0	n=1	n=2	n=3	n=4	Na	Al	Si	
		% of Si with n number of coordinated Al atoms						[wt%]	[wt%]	
NaY	2.6	8.9	38.1	41.9	11.1	0	8.3	4.5	28.2	
ENaY	2.2	5.7	29	46.3	16.4	2.6	7.8	4.2	23.1	
					4					
		$\Sigma \mathbf{I}_{\mathbf{Si} (\mathbf{n} \mathbf{Al})}$								
		Si			n = 0	n = 0				
		R = =								
		Al			4					
					Σ 0.2	$\Sigma = 0.25 \text{ n } \mathbf{I}_{Si (n Al)}$				
		n = 0								

Table S1 Deconvolution of the <sup>29</sup>Si NMR spectra and elemental analysis by ICP

The overall Si/Al ratio of the framework is estimated based on the above equation, where I is the intensity of the peak that is associated with n number of coordinated Al atoms.

Table S2 Elemental analysis by ICP

Catalyst	Са	к	Na	
	[wt%]	[wt%]	[wt%]	
NaY	-	-	8.3	
ENaY	-	-	7.8	
Ca-ENaY	3.1	-	6.9	
KOH-Ca-ENaY	3.7	3.7	7.2	
KOH-Ca-ENaY spent	3.7	2.7	7.4	
KOH-Ca-ENaY regen4x	3.6	3.7	7	

Table S3 Fitting of Ca k-edge XANES spectra

Catalyst	fresh Cat	Ca lactate	CaCO₃	KOH-Ca-ENaY@700	Sum
KOH-Ca-ENaY spent	66	34	0	0	100
KOH-Ca-ENaY regen 1x	24	0	52	24	100
KOH-Ca-ENaY regen 4x	2	0	66	32	100
KOH-Ca-NaY regen4x	0	0	74	26	100