

#### Figure captions

Fig. S1. MS signals of CO<sub>2</sub> (—) and CO (---) over 20FeAl-N catalyst.

Fig. S2. IR spectra of 20FeAl catalysts with different precursor: (a) 20FeAl-SA (III), (b) 20FeAl-SA (II), (c) 20FeAl-SA (II) after reaction for 3 h, (d) 20FeAl-SA (II) after reaction for 24 h.

Fig. S3. H<sub>2</sub>-TPR profiles of (a) 20FeAl-N; (b) 20FeAl-SA (III); (c) 20FeAl-SA (II); (d) 20FeAl-560-SO<sub>2</sub>.

Fig. S4. Py-FT-IR spectra of (a) 20FeAl-N, (b) 20FeAl-560-SO<sub>2</sub>, (c) 20FeAl-SA (II).

Fig. S5. Reaction results over (a) 20FeSi-SA (II), (b) 20FeZr-SA (II), (c) pure Al<sub>2</sub>O<sub>3</sub> in the reactant consisting of N<sub>2</sub>, SO<sub>2</sub> and C<sub>3</sub>H<sub>8</sub> at a molar ratio of 19:1:30 (Reaction conditions: T=560 °C, P=1 atm, F<sub>C<sub>3</sub>H<sub>8</sub></sub>=12 mL/min).

Fig. S6. Al 2p spectra of unsulfated and sulfated Fe<sub>2</sub>O<sub>3</sub>/Al<sub>2</sub>O<sub>3</sub> catalysts.

Fig. S7. Propane conversion (■), propylene yield (●) and methane yield (▲) over 20FeAl-N after reaction for 3 h, then: (a) introduced SO<sub>2</sub> at a molar ratio of N<sub>2</sub>: SO<sub>2</sub>: C<sub>3</sub>H<sub>8</sub>=19:1:30; (b) pre-treated with SO<sub>2</sub> for 3 h (Reaction conditions: T=560 °C, P=1 atm, F<sub>C<sub>3</sub>H<sub>8</sub></sub>=12 mL/min).

Fig. S8. MS signal of CO<sub>2</sub> during the reaction-regeneration cycles over 20FeAl-SA (II) catalyst.

Fig. S9. Effect of reaction-regeneration-sulfuration cycles on the performance of 20FeAl-N (Reaction conditions: T=560 °C, P=1 atm, F<sub>C<sub>3</sub>H<sub>8</sub></sub>=12 mL/min).

#### Table captions

Table S1. Reaction results of the blank experiment as well as the reaction over pure Al<sub>2</sub>O<sub>3</sub> (Reaction conditions: T=560 °C, P=1 atm, F<sub>C<sub>3</sub>H<sub>8</sub></sub>=12 mL/min).

Table S2. SO<sub>2</sub>-TG results at different temperature.

Table S3. The areas of reduction peaks quantified by peak deconvolution for sulfated and

unsulfated catalysts

Table S4. S 2p peak fitting calculation and analytical data of surface content (Fe, Al and S).

Table S5. Textural properties of fresh, reacted, regenerated 20FeAl-SA (II) and 20FeAl-560-SO<sub>2</sub>.

Table S1

Reaction results of the blank experiment as well as the reaction over pure Al<sub>2</sub>O<sub>3</sub> (Reaction conditions: T=560 °C, P=1 atm, F<sub>C<sub>3</sub>H<sub>8</sub></sub>=12 mL/min).

Catalyst	Conversion, wt%	Yield of propylene, wt%	Yield of methane, wt%
Blank	1.13	0.4	0.15
Al <sub>2</sub> O <sub>3</sub>	1.05	0.47	0.10

Table S2

SO<sub>2</sub>-TG results at different temperature

T, °C	Weight increase rate, wt%
450	19.1
560	11.9
600	8.6
Al <sub>2</sub> O <sub>3</sub> -700	5.5
Fe <sub>2</sub> O <sub>3</sub> -700	2.8

Table S3

The areas of reduction peaks quantified by peak deconvolution for sulfated and unsulfated catalysts

Catalyst	Peak 1	Peak 2'	Peak 2	Peak 3
20FeAl-N	2695.7	--	2204.8	13627.2
20FeAl-SA (III)	40.5	--	6917.5	10923.8
20FeAl-SA (II)	76.9	--	12386.4	9456.8
20FeAl-560-SO <sub>2</sub>	134.0	1702.5	4531.9	10994.8

Table S4

S 2p peak fitting calculation and analytical data of surface content (Fe, Al and S)

Catalyst	E <sub>b</sub> , eV	% area	Surface content of S, %
20FeAl-560-SO <sub>2</sub>	169.8	-	1.6
20FeAl-SA (II)	169.8	-	2.2
20FeAl-SA (II) after reaction for 3h	168.9 162.5	57.5 42.5	0.8
20FeAl-SA (II) after reaction for 24h	168.7 162.4	51.1 48.9	0.8
20FeAl-SA (III) after reaction for 24h	168.5 161.7	44.5 55.5	0.9
20FeAl-560-SO <sub>2</sub> after reaction for 6 h	168.9 162.2	62.2 37.8	1.0

Table S5

Textural properties of fresh, reacted, regenerated 20FeAl-SA (II) and 20FeAl-560-SO<sub>2</sub>

Catalysts	S <sub>BET</sub> , m <sup>2</sup> /g	Pore diameter, nm	Particle volume, cm <sup>3</sup> /g
Fresh 20FeAl-SA (II)	125.4	6.53	0.263
20FeAl-SA (II) after reaction for 24 h	108.3	5.62	0.170
Regenerated once 20FeAl- SA (II)	117.0	6.50	0.258
Fresh 20FeAl-560-SO <sub>2</sub>	149.1	6.60	0.258
Reacted (one cycle) 20FeAl- 560-SO <sub>2</sub>	140.8	5.65	0.249
Regenerated once 20FeAl- 560-SO <sub>2</sub>	145.8	6.58	0.254
Reacted (four cycle) 20FeAl-560-SO <sub>2</sub>	136.9	6.44	0.245
Regenerated four times 20FeAl-560-SO <sub>2</sub>	142.2	6.50	0.249

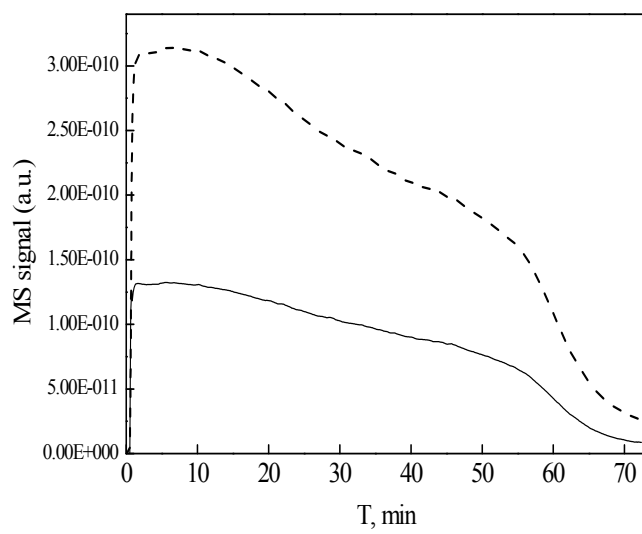


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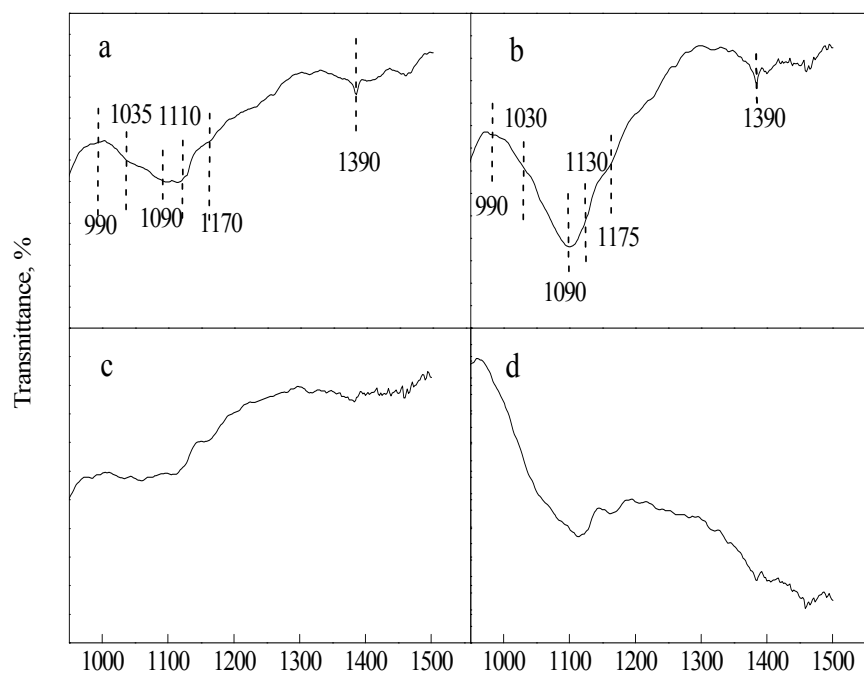


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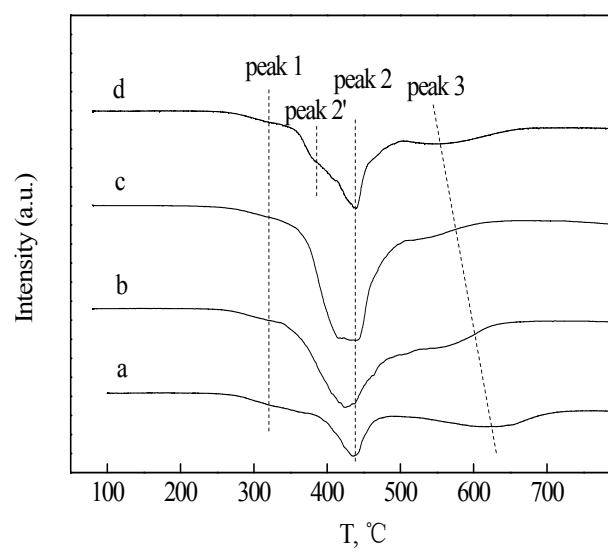


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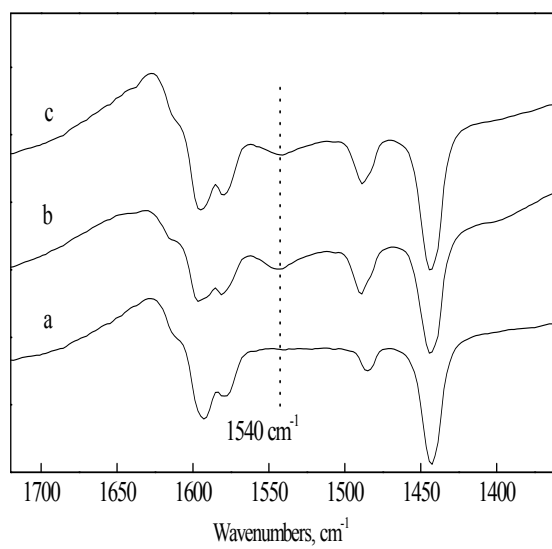


Fig. S4. Py-FT-IR spectra of (a) 20FeAl-N, (b) 20FeAl-560-SO<sub>2</sub>, (c) 20FeAl-SA (II) (desorption at 200 °C).



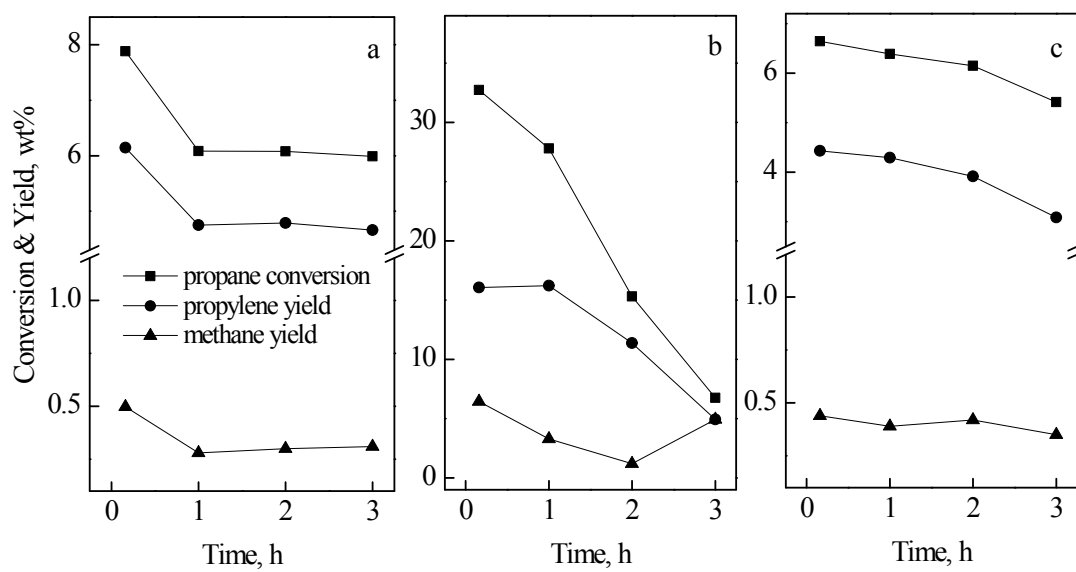


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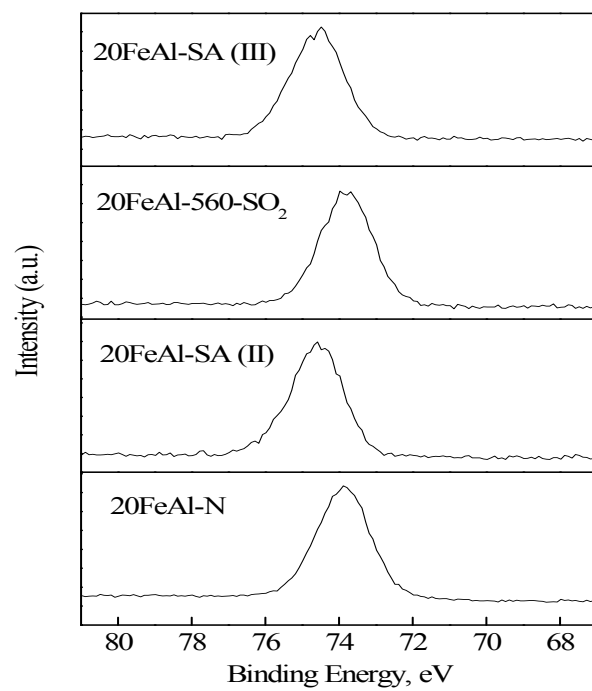


Fig. S6. Al 2p spectra of unsulfated and sulfated Fe<sub>2</sub>O<sub>3</sub>/Al<sub>2</sub>O<sub>3</sub> catalysts.

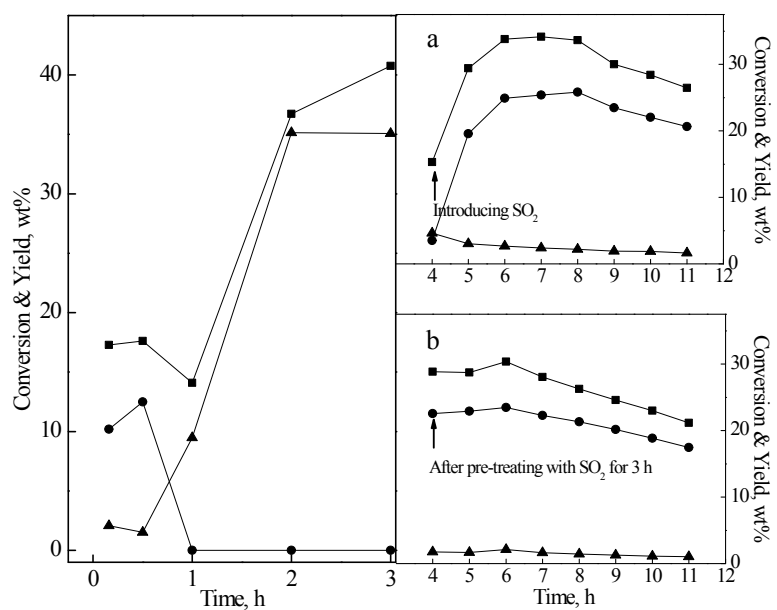


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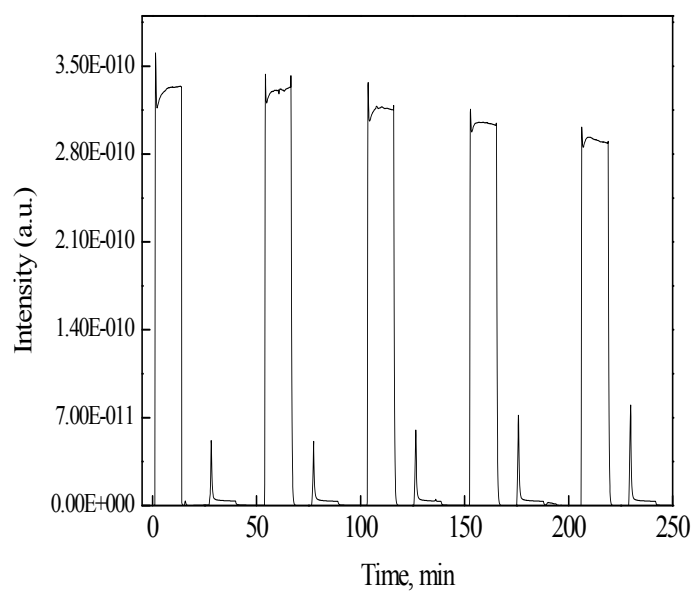


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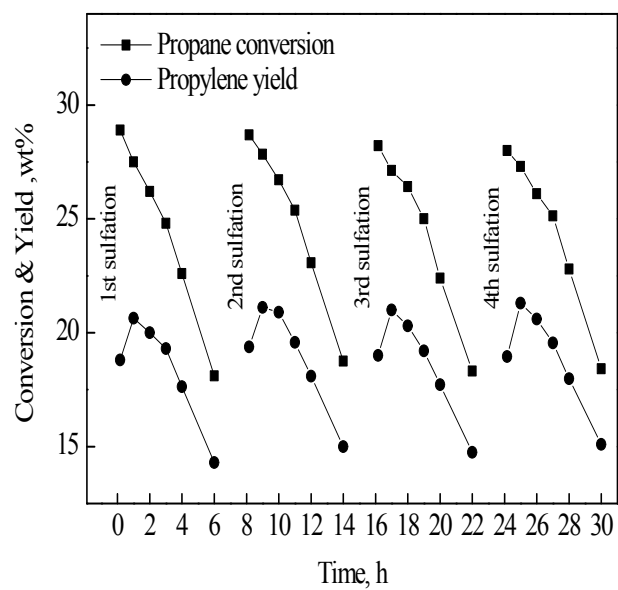


Fig. S9. Effect of reaction-regeneration-sulfuration cycles on the performance of 20FeAl-N (Reaction conditions: T=560 °C, P=1 atm,  $F_{C_3H_8}$ =12 mL/min).