

Supplementary Information:

One-pot synthesis of N-methylpyrrolidine (NMPD) using Cu- and Ni- modified ZSM-5 as an efficient catalyst

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General procedures for the preparation of 3%Cu-3%Ni/ZSM-5

The 3%Cu-3%Ni/ZSM-5 catalyst was prepared by incipient wetness method using $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$ and $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ as starting materials. 1.14g $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$ (0.3g Cu) and 1.5g $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (0.3g Ni) dissolved in 3mL water, then 10g HZSM-5 was added zeolite with continuous stirring at room temperature. After 2h stirring and 2h aging, the resulted precipitate was dried at 110 °C for 12h. Then, the precursors were calcined at 500 °C for 3h in air. The calcined catalysts were denoted as 3%Cu-3%Ni/ZSM-5.

Table S1 The contents of Cu and Ni measured by AAS

| catalyst | Content wt% | |
|---------------------|-------------|------|
| | Cu | Ni |
| 1.5%Cu-1.5%Ni/ZSM-5 | 1.43 | 1.47 |
| 3%Cu-3%Ni/ZSM-5 | 2.92 | 2.89 |
| 2%Cu-4%Ni/ZSM-5 | 2.36 | 3.93 |
| 3%Cu/ZSM-5 | 2.87 | |
| 6%Ni/ZSM-5 | | 5.52 |
| 3%Cu-3%Ni/ZSM-5* | 2.14 | 2.59 |

* after 5 cycles

Table S2 BET analyses of different samples

| Sample | Specific surface area (m^2/g) | Pore volume (cm^3/g) | Average pore radius (nm) |
|------------------------------|---|--|--------------------------|
| HZSM-5 | 388 | 0.355 | 2.459 |
| 3%Cu-3%Ni/ZSM-5 | 285 | 0.249 | 2.348 |
| 6%Cu-6Ni%/ZSM-5 | 260 | 0.205 | 2.252 |
| 3%Cu-3%Ni/ZSM-5 ^a | 281 | 0.182 | 2.163 |
| 3%Cu-3%Ni/ZSM-5 ^b | 299 | 0.276 | 2.215 |

^a the Si/Al ratio was 50; ^b the Si/Al ratio was 300

Table S3 XPS peak table of 3%Cu-3%Ni/ZSM-5

| Name | Start BE | Peak BE | End BE | Atomic % |
|------|----------|---------|--------|----------|
| C1s | 2980.8 | 284.8 | 279.28 | 93.33 |
| Cu2p | 970.08 | 932.98 | 925.28 | 3.13 |
| Ni2p | 890.08 | 855.8 | 845.28 | 3.54 |

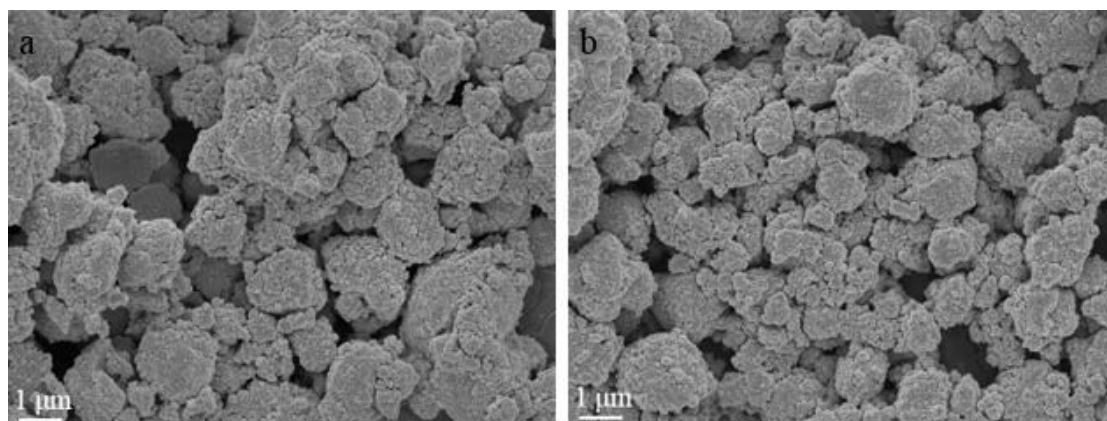


Fig. S1 SEM images of a) H-ZSM-5, b) 3%Cu-3%Ni/ZSM-5

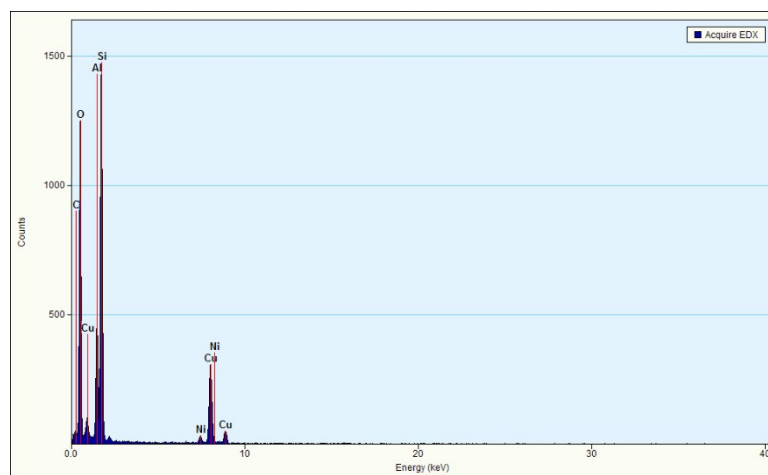


Fig. S2 the EDX pattern of 3%Cu-3%Ni/ZSM-5

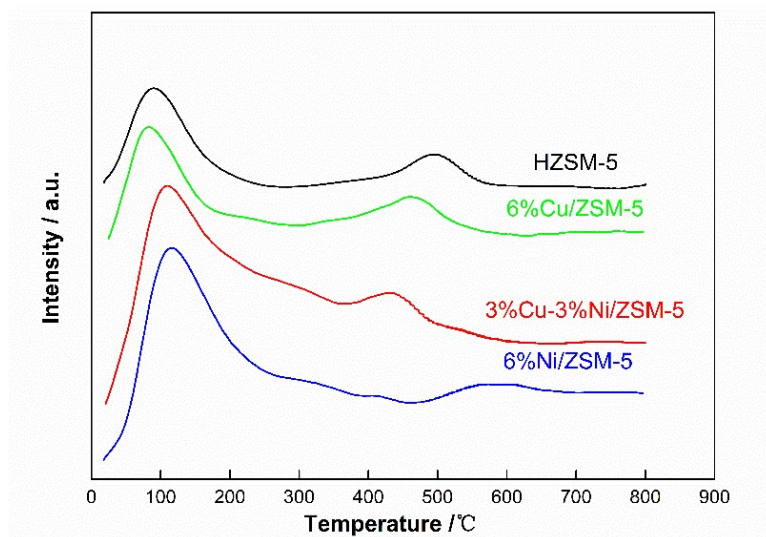


Fig. S3 The TPD curves of different catalysts