# ACS APPLIED NANO MATERIALS

Irregularly Shaped NiO Nanostructures for Catalytic Lean Methane Combustion

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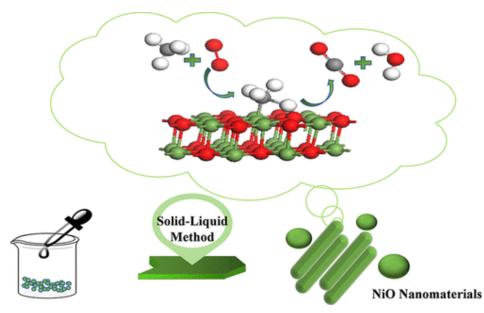
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### SUBJECTS:

<u>Catalysts</u>, <u>Hydrocarbons</u>, <u>Oxides</u>, <u>Precipitation</u>, Redox reactions

### Abstract



NiO nanomaterials prepared using a solid-liquid NH<sub>3</sub>·H<sub>2</sub>O precipitation method (NiO-NSL) were tested in the catalytic combustion of methane. The NiO-NSL presented a characteristic rod-like nanostructure with a length of about a few hundred nanometers except for a part of the nanoparticles. For comparison, the NiO nanomaterials prepared by the traditional liquid-phase  $NH_3 \cdot H_2O$  precipitation method (NiO-NLL) were tested in the same reaction conditions. NiO-NSL exhibited significantly higher methane combustion activity than NiO-NLL and achieved the complete combustion of methane at 390 °C, which was outstanding in non-noble metal-based catalyst. X-ray photoelectron spectroscopy (XPS) and hydrogen-temperature-programmed reduction  $(H_2-TPR)$  results indicate that the surface Ni<sup>2+</sup> content of NiO-NSL was higher than that of NiO-NLL, and the presence of more  $Ni^{2+}$  might be responsible for the enhanced activity. DFT calculations prove that the energy barrier for C-H bond activation on Ni<sup>2+</sup> was lower than that on Ni<sup>3+</sup>, which was consistent with the higher methane catalytic combustion activity of NiO-NSL. In addition, when the precipitating agent was replaced with NaOH and (NH4)<sub>2</sub>CO<sub>3</sub>, the generalization of the solid-liquid precipitation method in the preparation of the NiO catalysts was also tested. The results show that the solid-liquid precipitation method proposed in this work was still applicable when NaOH was used as a precipitant. However, with the use of  $(NH_4)_2CO_3$  as a precipitant, the methane catalytic activity of the NiO nanoparticles prepared by the solid-liquid precipitation method was reduced to a certain extent compared with the traditional liquid-phase precipitation method. This research can open up a highly efficient and environmentally friendly method for the synthesis of methane combustion catalysts.

### KEYWORDS:

solid-liquid precipitation method

rod-like nanostructure

methane catalyticShow More

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Stability test results of NiO-NSL (Figure S1); wide-angle XRD patterns of the as-precipitated samples using  $(NH_4)_2CO_3$  as the precipitant and a standard  $xNiCO_3 \cdot yNi(OH)_2$  sample (Figure S2); representative TEM images of the precursors and as-calcined catalysts of the other four samples (Figures S3 and S4); specific performance data of samples in previous research (Table S1); XPS analysis of the other samples (Table S2, Figures S5 and S6); H<sub>2</sub>-TPR profiles of the other samples (Table S3, Figures S7 and S8); detailed DFT calculation results (Table S4); the surface model used in the calculations (Figure S9) (PDF)

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S-3
Figure S2.
Wide-angle XRD patterns of the as-precipitated samples using
(NH
4
)
2

```
СО
3
 as precipitant and standard xNiCO
3
·yNi(OH)
2
 sample.
They
both showed typical crystal structure of monoclinic
nullaginite (JCPDS
PDF#35-0501). The crystallinity of xNiCO
3
·yNi(OH)
2
-NCSL was slightly
better than that of xNiCO
3
·yNi(OH)
2
-NCLL since the diffraction peaks of
xNiCO
3
·yNi(OH)
2
-NCSL were slightly sharper than those of the latter.
S-4
Figure S3.
  Representative TEM images of (a) Ni(OH)
2
-NaLL,
       (b)
Ni(OH)
2
-NaSL,
```

```
(C)
xNiCO3 · yNi (OH) 2-NCLL
and
(d)
xNiCO3 ·yNi(OH)2-NCSL.
Figure S4.
Representative TEM images of (a) NiO-NaLL, (b) NiO-NaSL,
(c) NiO -NCLL and (d) NiO -NCSL.
S-5
Figure S5.
 X-ray photoelectron spectroscopy (XPS) results of (a) Ni
2p
and (b) O 1s spectra of NiO-NaSL and NiO-NaLL.
S-6
Figure S6.
 X-ray photoelectron spectroscopy (XPS) results of (a) Ni
2p
and (b) O 1s spectra of NiO-NaSL and NiO-NaLL.
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