

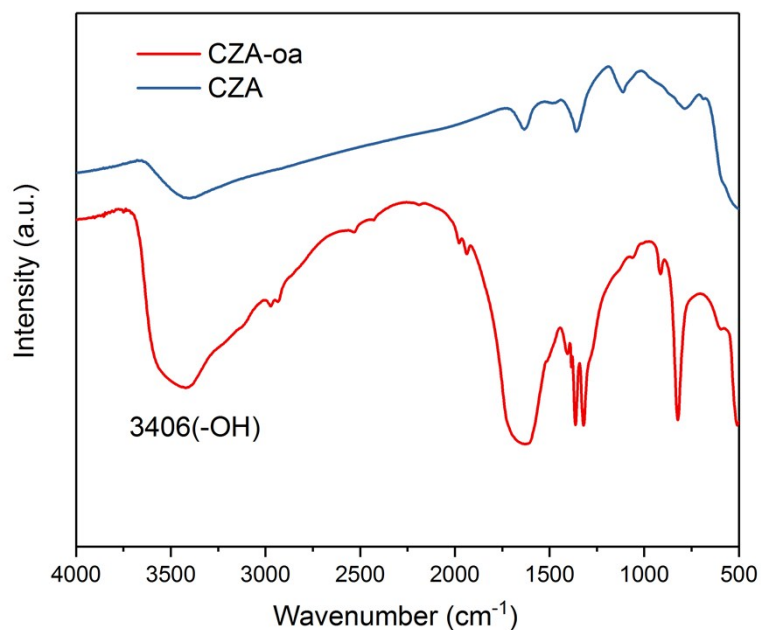
## Electronic Supplementary Information

### Direct coating copper-zinc-aluminum oxalate with H-ZSM-5 to fabricate highly efficient capsule bifunctional catalyst for dimethyl ether production from syngas

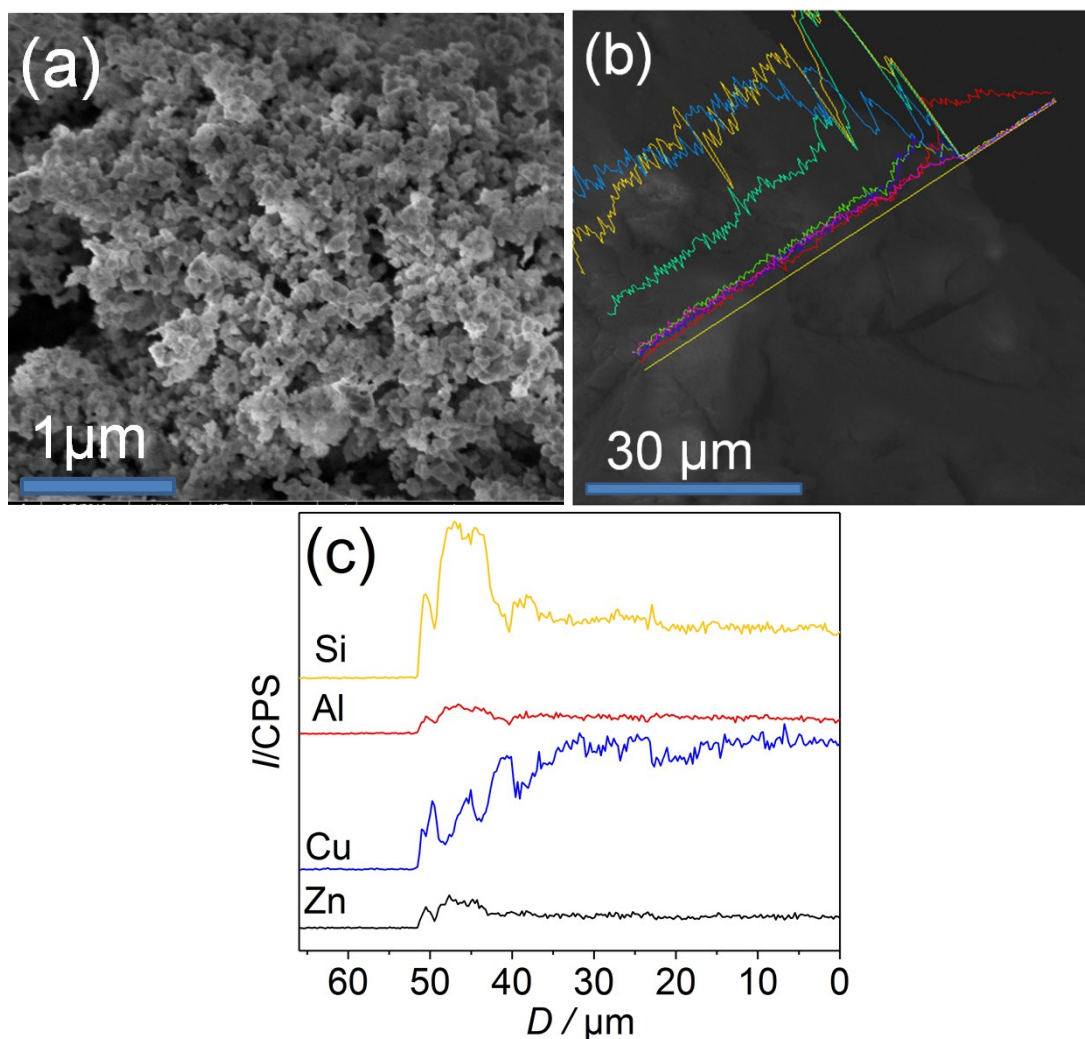
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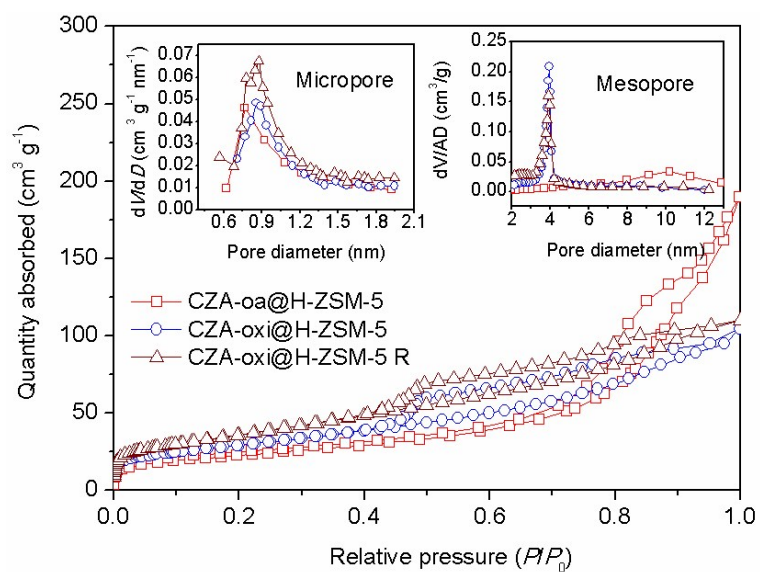


**Fig. S1** FT-IR spectra of copper-zinc-aluminum oxalate (CZA-ox) and the copper-zinc-aluminum oxide (CZA-oxi) prepared by calcining the oxalate CZA-ox at 330 °C for 2 h in air.

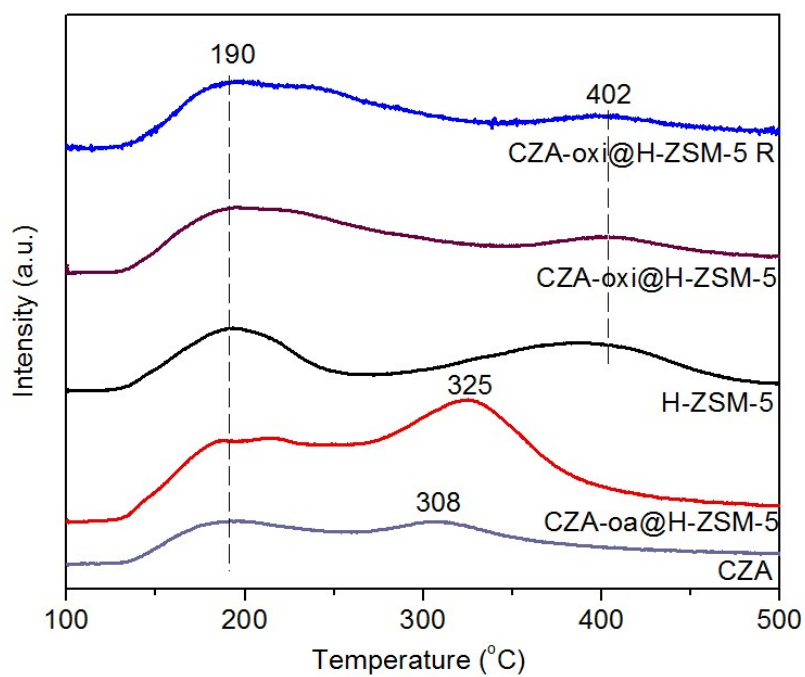


**Fig. S2** (a) Surface SEM image, (b) cross-section SEM image with EDS scanning, and (c) EDS line analysis of the CZA-oxi@H-ZSM-5 R capsule bifunctional capsule catalyst prepared according to reference.

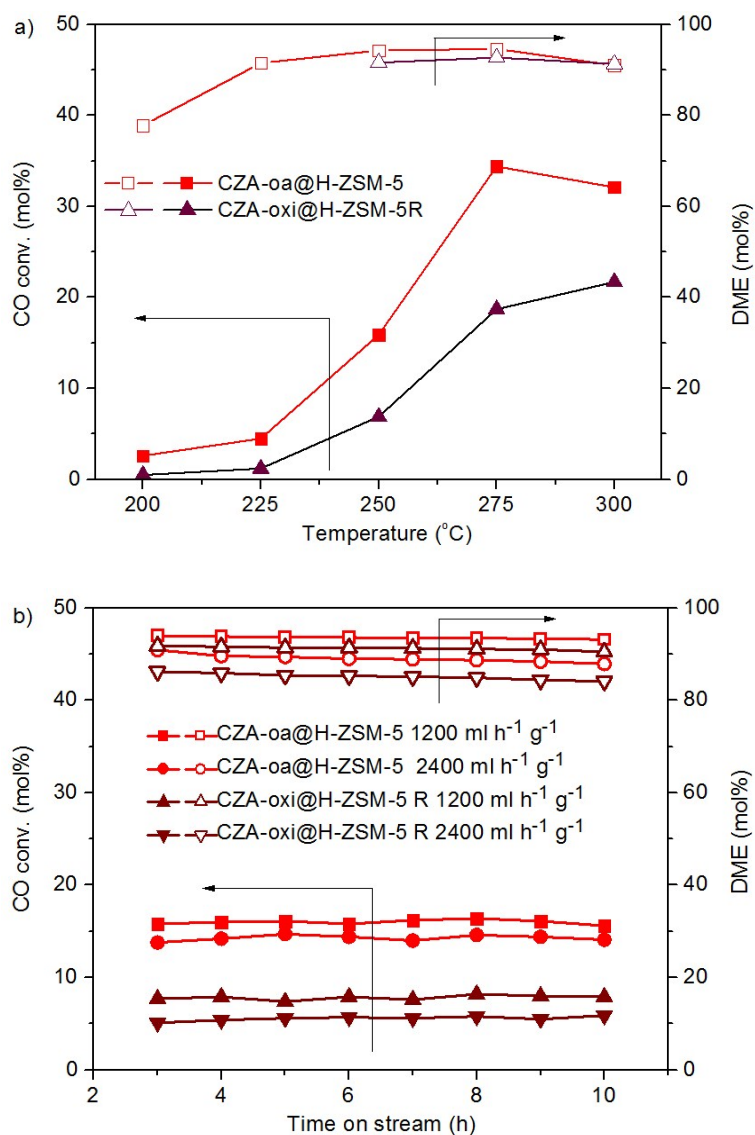
Scanning electron microscopy (SEM) images of CZA-oxi@H-ZSM-5 R catalyst are shown in Figure S2. It can be found that the CZA core is covered by a uniform layer of zeolite, and line scan is performed along the mark in the Figure S2b to obtain a structure similar to CZA-oxi@H-ZSM-5. When transitioning from the zeolite layer to the core catalyst, the signal intensity of Si is drastically reduced, and the signals of Cu and Zn are significantly enlarged, suggesting the successful coating. In addition, the zeolite shell thickness of 7  $\mu\text{m}$  is confirmed.



**Fig. S3** N<sub>2</sub> adsorption-desorption isotherms of the as-synthesized CZA-*oa*@H-ZSM-5, CZA-*oxi*@H-ZSM-5, and CZA-*oxi*@H-ZSM-5 R bifunctional capsule catalysts. Inset: Micropore size distribution from H-K method and Mesopore size distribution by BJH method from adsorption branch.



**Fig. S4** NH<sub>3</sub>-TPD profiles of the CZA-oa@H-ZSM-5, CZA-oxi@H-ZSM-5, and CZA-oxi@H-ZSM-5 R capsule catalysts, and H-ZSM-5 shell is included for comparison.



**Fig. S5** Catalytic performance in direct DME production from syngas over the developed CZA-oxa/HZSM-5 and the previously reported CZA-oxi/HZSM-5 R under the different reaction temperatures (a) and gas hourly space velocities (b).

**Table S1**Quantitative data of H<sub>2</sub>-TPR, XRD and NH<sub>3</sub>-TPD measurements.

catalyst	H <sub>2</sub> consumption	NH <sub>3</sub> uptake	CuO average
	(mmol g <sup>-1</sup> )	(μmol g <sup>-1</sup> )	crystalline size (nm)
CZA	2.48	85	11.4
CZA-oa@H-ZSM-5	1.72	170	20.5
CZA-oxi@H-ZSM-5	1.62	124	17.8
CZA-oxi@H-ZSM-5 R	1.36	120	14.3
H-ZSM-5	-	128	-

**Table S2**

XPS quantitative analysis of CZA, CZA-oa@H-ZSM-5 and CZA-oxi@H-ZSM-5 R.

Catalyst	Atomic ratio of Cu/Zn	XPS analysis Cu 2p3/2				XPS analysis Zn 2p1/2	
		Binding energy (eV)		Percentage (%)		Binding energy (eV)	
		Cu <sup>2+</sup>	Cu <sup>+</sup>	Cu <sup>2+</sup>	Cu <sup>+</sup>	Peak α	Peak β
CZA	0.81:1	934.3	932.7	40.0	60.0	1044.7	1040.1
CZA-oa@H-ZSM-5	0.77:1	934.4	932.9	45.4	54.6	1044.9	1040.3
CZA-oxi@H-ZSM-5 R	0.64:1	934.4	932.8	56.2	43.8	1044.9	1039.5

**Table S3**

The DME formation rate per m<sup>2</sup> of exposed Cu atoms and per g catalyst of CZA-*oa*@H-ZSM-5, CZA-*oxi*@H-ZSM-5, and CZA-*oxi*@H-ZSM-5 R catalysts.

catalyst	$R_{\text{DME}}^a$ (mmol m <sup>-2</sup> <sub>Cu</sub> h <sup>-1</sup> )	$R'_{\text{DME}}^b$ (mmol g <sup>-1</sup> <sub>cat</sub> h <sup>-1</sup> )
CZA- <i>oa</i> @H-ZSM-5	0.122	1.09
CZA- <i>oxi</i> @H-ZSM-5	0.048	0.51
CZA- <i>oxi</i> @H-ZSM-5 R	0.031	0.53

<sup>a</sup>DME formation rate per m<sup>2</sup> of exposed Cu atoms and h on the catalysts. <sup>b</sup>DME productivity per g catalyst and h.