### **Support information**

## **Regeneration Mechanism of Deactivated Zeolite-Supported Catalyst for the Combustion of Chlorinated Volatile Organic Compounds**

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The semi-quantitative ratio of Bronsted acid sites to Lewis acid sites[1]: as C (pyridine on B sites) = 1.88 IA(B) R2/W and C(pyridine on L sites) =  $1.42 \text{ IA(L)} \text{ R2/W}(\text{C} \text{ is the concentration (mmol g-1 catalyst), IA (B, L) is the integrated absorbance of the B or L band (cm<sup>-1</sup>), R is the radius of the catalyst disk (cm), and W is the weight of the disk (mg)).$ 

 Emeis C: Determination of integrated molar extinction coefficients for infrared absorption bands of pyridine adsorbed on solid acid catalysts. *Journal of Catalysis* 1993, 141(2):347-354.

#### 3.2 Morphology characterization



Fig.S1 The EDS-Mapping test of used CNH

#### 3.3 Sediment deposition identification



Fig.S2 The  $H_2$ -TPR profiles of used CNH and CNH- $T_{DRY}$ 

For fresh CNH, the peak at 202 °C was reported originating from highly disperse amorphous CuO species[2]. And peaks at 225°C and 255°C was due to reduction temperature of Cu<sup>2+</sup> $\rightarrow$ Cu<sup>+</sup> and Cu<sup>+</sup> $\rightarrow$ Cu<sup>0</sup>, which was promoted by Nb-Cu-O bimetallic bonding[3]. The peak at 585 °C was ascribed to the reduction of surface Nb<sub>2</sub>O<sub>5</sub>. For used CNH and CHN-T<sub>DRY</sub>, it can be found that temperature and amount of reduction peaks for Cu<sup>2+</sup> $\rightarrow$ Cu<sup>+</sup> (230°C) and Cu<sup>+</sup> $\rightarrow$ Cu<sup>0</sup>(230°C) for CNH-T<sub>DRY</sub> are obviously better than that of used CNH, which proved that Nb-Cu-O

bimetallic bonding has formed during regeneration process and promote the reduction ability of CuO[3]. For CNH-T<sub>WET</sub>, the reduction of CuO<sub>x</sub> has obvious decreased, which is due to the high humidity water environment leads to the formation of Cu(OH)<sub>2</sub>.

[2]J.H. Kwak, R.G. Tonkyn, D.H. Kim, J. Szanyi, C.H. Peden, Journal of Catalysis 275 (2010) 187.

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Fig. S3 The abundance spectrums for detected chemical compound by GC/MS.



Fig. S4 Transmission infrared spectrum of CNH, used CNH, CNH- $T_{DRY}$ , and CNH- $T_{WET}$ .



### 3.4 Mechanism of surface coke deposition

Fig. S5 In situ DRIFT spectra of surface reaction on catalyst CNH-T<sub>DRY</sub> collected at (a) 250 °C and (b) 400 °C.

# 3.5 Surface property



Fig. S6 The XPS spectra of Cu LMM for fresh, aged, regenerated catalysts.

#### **Density Functional Theory (DFT) calculations:**

All calculations were performed within the framework of DFT as implemented in Vienna Ab initio Software Package (VASP 5.3.5) code within the Perdew–Burke– Ernzerhof (PBE) generalized gradient approximation and the projected augmented wave (PAW) method [4, 5]. The cut-off energy for plane-wave basis set was set to 400 eV. A 2 × 3 supercell of CuO(101) surface including 5 layers; and a 1 × 1 supercell of the Nb<sub>2</sub>O<sub>5</sub> (001) surface including 5 layers; were constructed to model the catalyst structure in this work. A vacuum layer of 12 Å was introduced to avoid interactions between periodic images. The Brillouin zone of surface unit cell was sampled by Monkhorst–Pack (MP) grids, with different k-point meshes for CuO and Nb<sub>2</sub>O<sub>5</sub> structure optimizations [6]. The CuO and Nb<sub>2</sub>O<sub>5</sub> surfaces were determined by  $3 \times 2 \times 1$  and  $4 \times 1 \times 1$  MP grid. The convergence criterion for electronic self-consistent iteration and force was set to 10-5 eV and 0.01 eV/Å, respectively. The climbing image nudged elastic band (CI-NEB) [7,8] method was used to confirm the transition states with only one imaginary frequency along the reaction coordinates.

The adsorption energy (Eads) of surface species is defined by:

Eads = Etotal - Esurface - Sspecies

where Etotal represents the total energy of adsorbed species on catalyst surface, Esurface is the energy of empty surface, and Especies is the energy of species in the gas phase.

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