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Novel efficient U-Cu/CSAC catalyst for selective reduction of NO at low temperatures: Preparation, characterization, evaluation and mechanism

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Electronic Supplementary Information

Fig. S1 The image of raw industrial-grade CSAC.

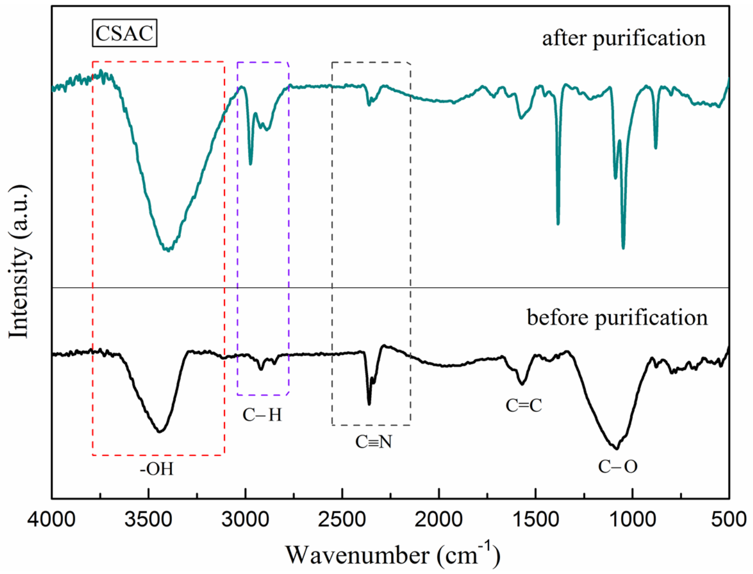

Fig. S2 The FT-IR spectra of CSAC carrier before and after purification.

Band assignments:[S1–S6]

The peak at 3445 cm^{-1}: the hydroxyl group (-OH). It was attributed to the stretching vibration of O–H in the range of 3450–3442 cm^{-1};

The peak between 2970–2880 cm^{-1}: C–H;

The peak at 2370 cm^{-1}: the isonitrile C≡N cyano terminal group;

The peak between 1695–1630 cm^{-1}: C=C;

The peak between 1125–1090 cm^{-1}: C–O. It was attributed to the stretching vibration of C–O in the range of 1125–1090 cm^{-1}

The description of Fig. S2 was already shown in the main text.

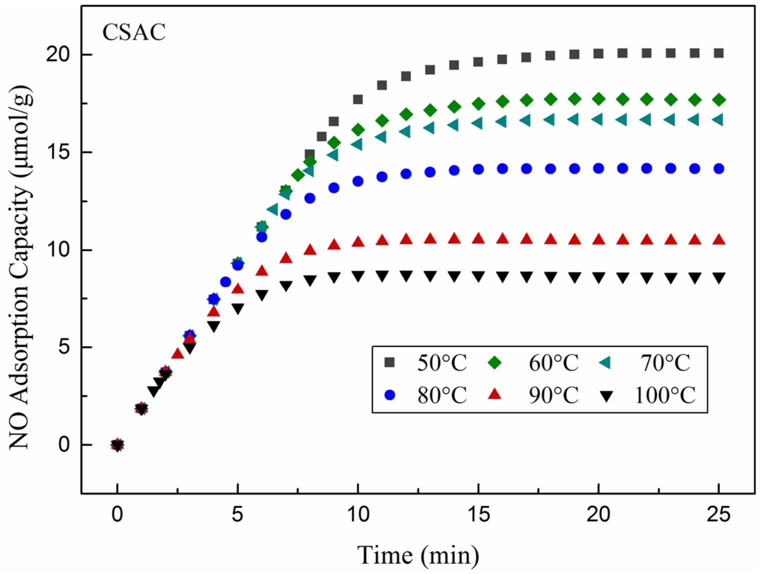

Fig. S3 The adsorption capacity of CSAC carrier at different temperatures.

The NO adsorption capacity at different temperatures was calculated by the following formula,

$$q = \frac{1}{M}\left[\int_0^t Q\,\frac{c_0 - c}{1 - c}\,dt\right]\frac{T_0}{T}\,\frac{1}{V_m}$$

q is the NO adsorption capacity of unit mass activated carbon;

M is the mass of AC carrier;

t is the adsorption time;

Q is the inlet gas flow;

c_0 is the NO concentration at the inlet;

c is the NO concentration at the exit;

T_0 is 273 K;

T is the adsorption temperature;

V_m is 22.4 mL/mmol.

Concretely, the NO adsorption capacity of CSAC carrier at 50, 60, 70, 80, 90 and 100°C were 20.1, 17.7, 16.7, 14.2, 10.5 and 8.6 μmol/g, respectively.

Table S1 The chemical reactions involved in the processes of low-temperature urea-SCR and their standard Gibbs free energy changes.

Reaction	ΔG_T^θ (kJ)*	NO.
$2NO + 2/3CH_4N_2O \rightarrow 5/3N_2 + 2/3CO_2 + 4/3H_2O$	−625.8 ~ −614.8	(1)
$2NO + O_2 \rightarrow 2NO_2$	−59.7 ~ −67.1	(2)
$2NO_2 + 4/3CH_4N_2O \rightarrow 7/3N_2 + 4/3CO_2 + 8/3H_2O$	−1020.6~ −989.8	(3)
$2NO_2 + 2CH_4N_2O + O_2 \rightarrow 3N_2 + 2CO_2 + 4H_2O$	−1475.1~ −1432.0	(4)
$NO + NO_2 + CH_4N_2O \rightarrow 2N_2 + CO_2 + 2H_2O$	−823.2~ −802.3	(5)
$NO + NO_2 + 3/2O_2 + 2CH_4N_2O \rightarrow 3N_2 + 2CO_2 + 4H_2O$	−1505.0~ −1465.5	(6)

* ΔG_T^θ is the value of the Gibbs free energy change during 50~100°C

The standard Gibbs free energy changes (ΔG) for the six reactions were calculated by the following formula,

$$\Delta G = \Delta H - T\Delta S$$

ΔG is the standard Gibbs free energy change of reaction; ΔH is the enthalpy change of chemical reaction; ΔS is the entropy change of chemical reaction; T is the reaction temperature.

The description of Table S1 was already shown in the main text.

According to Wang and Shah [S7,S8], ΔG can be used as a tool to judge the spontaneous direction and difficulty degree of chemical reaction. For a chemical reaction, when the value of ΔG is negative, the reaction can occur spontaneously in the direction of positive. Besides, the larger the value of ΔG, the less likely the reaction is to proceed spontaneously [S8].

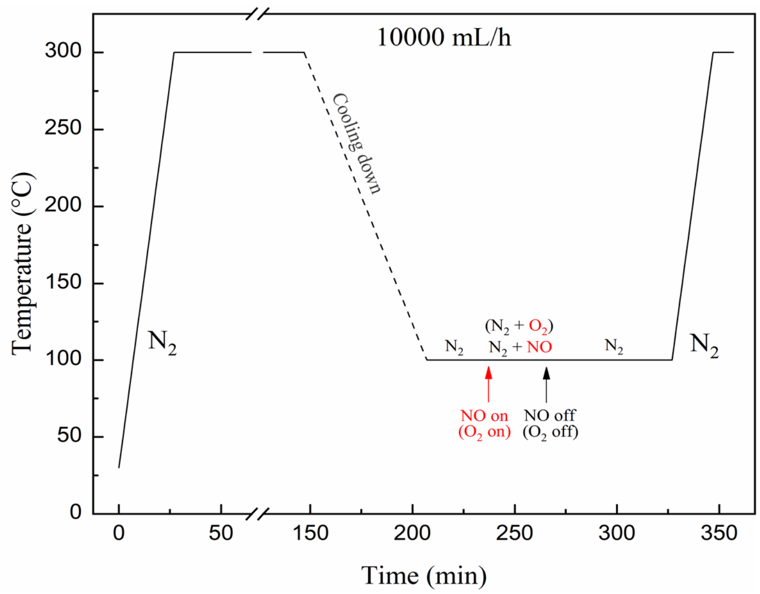

Fig. S4 The process of programmed temperature and experimental atmosphere changes.

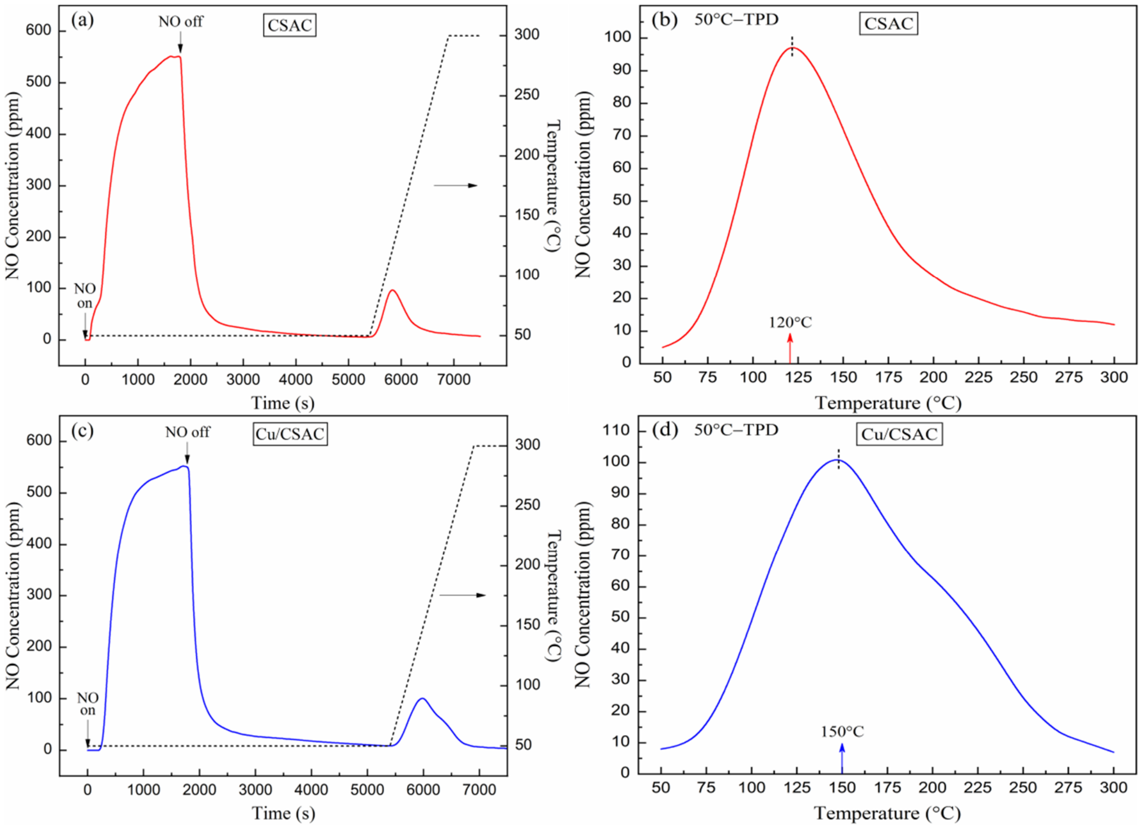

Fig. S5 The experimental results of NO adsorption-desorption and NO-TPD for CSAC carrier and Cu/CSAC catalyst at 50ºC.

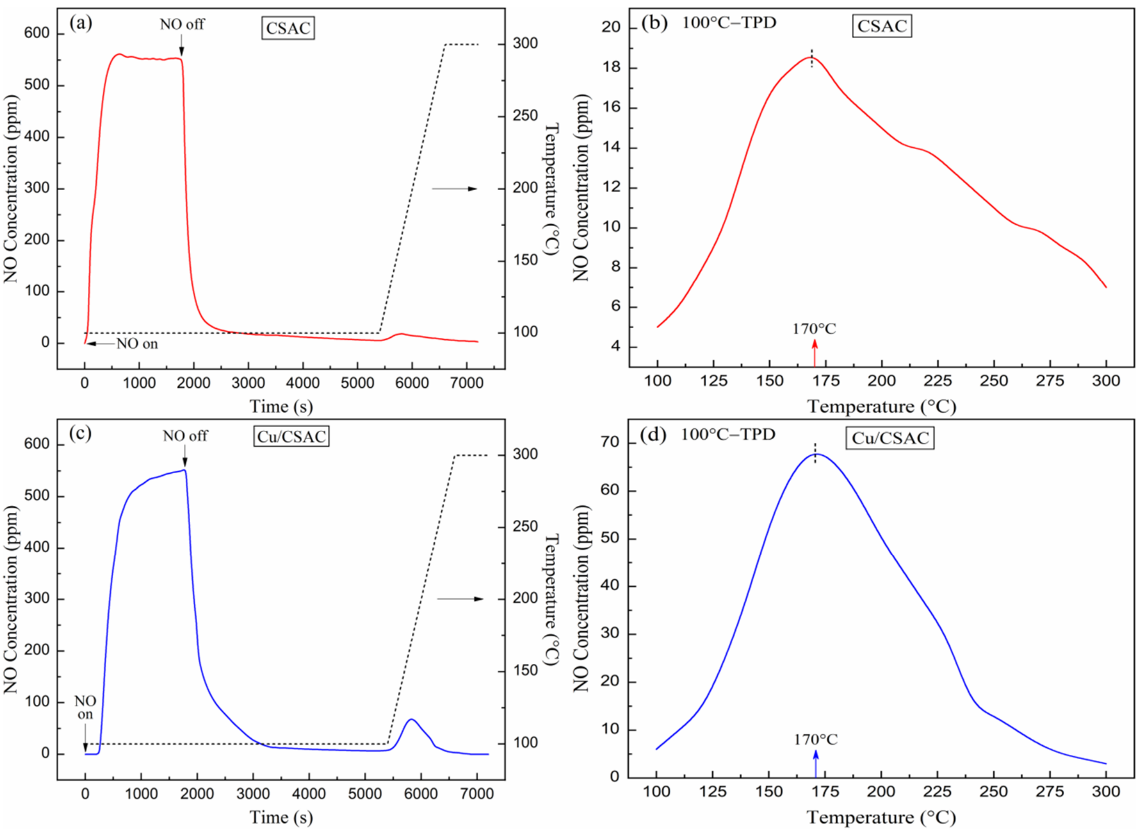

Fig. S6 The experimental results of NO adsorption-desorption and NO-TPD for CSAC carrier and Cu/CSAC catalyst at 100ºC.

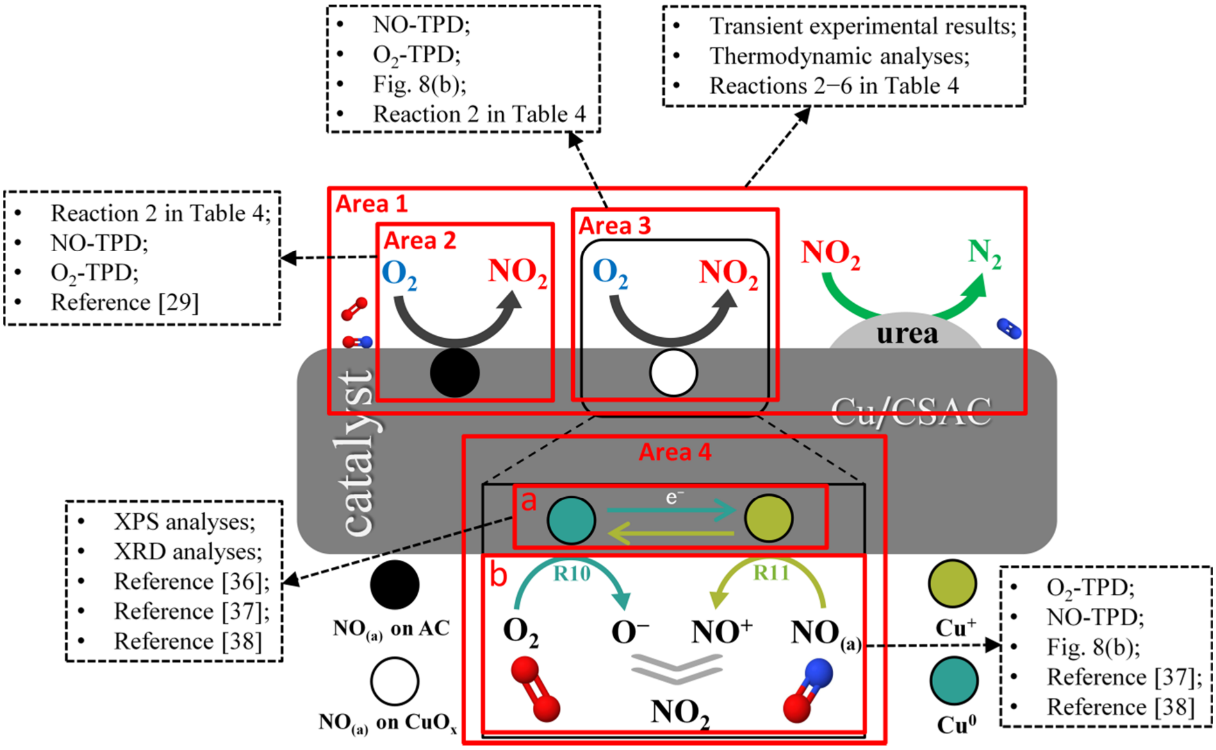

Fig. S7 The supporting information corresponding to each area of the mechanism diagram.

Additional Notes for Area 1: According to the analyses of transient experimental results (Fig. 10 in the manuscript) and thermodynamic analyses (Table 4 in the manuscript), it is known that the oxidation of NO (reaction (2) in Table 4) is the rate-limiting reaction in the process of LTU-SCR. Moreover, the transient experimental result also indicated that the ability of direct reduction of NO by urea in the absence of O_2 (the reaction (1) in Table 4) was very small (which has been mentioned in the main document). Area 1 represents the main reactions for LTU-SCR (NO is first oxidized to NO_2 and then reduced by the reducing agent; reactions 2−6 in Table 4), and the supporting data is the transient experimental results (Fig. 10) and thermodynamic analyses (Table 4).

References

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