Electronic Supplementary Material (ESI) for Catalysis Science & Technology.

Supporting information

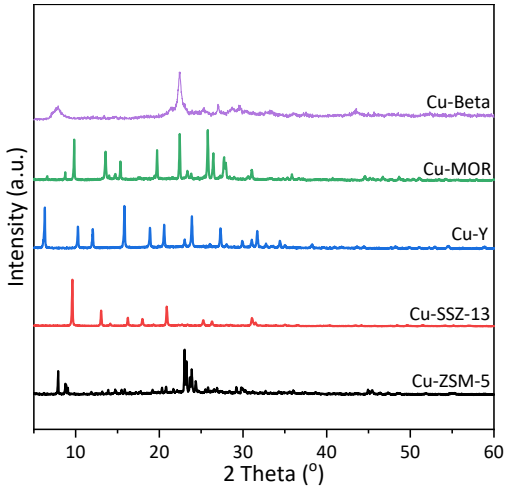

Fig. S1. XRD spectra of Cu based catalyst with different topology structure.

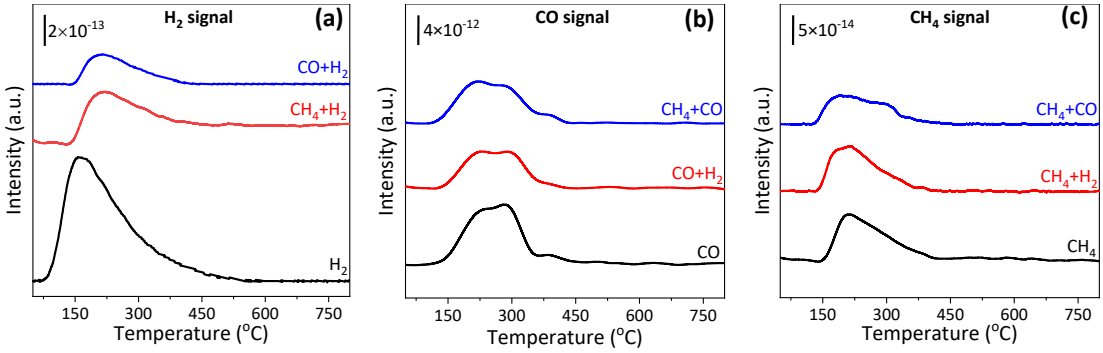

Fig. S2. H_2 signal (a), CO signal (b), CH_4 signal (c) of Cu-ZSM-5 catalyst at different adsorption conditions.

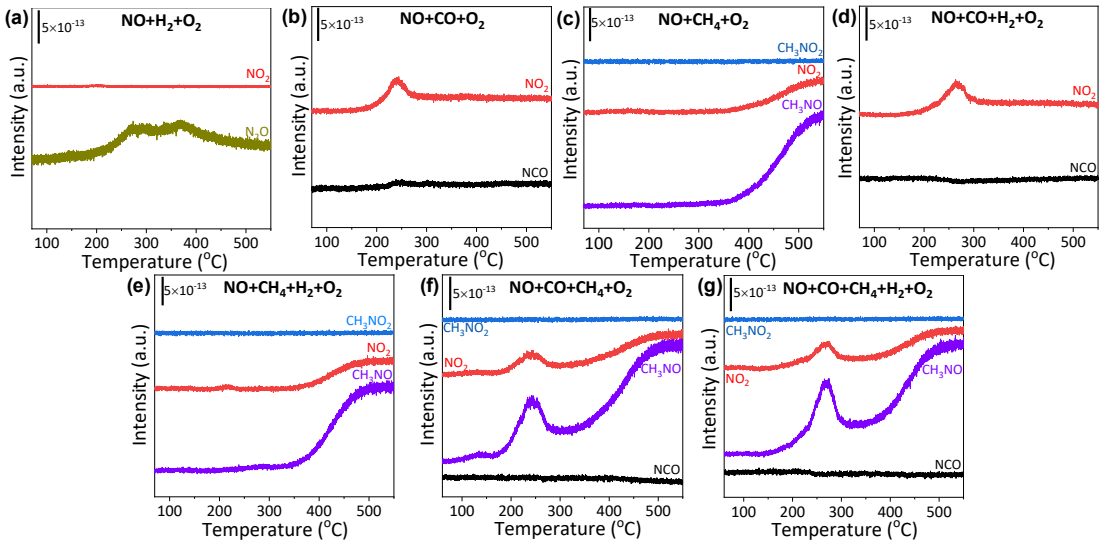

Fig. S3. MS signals of Cu-ZSM-5 catalyst at different reaction conditions.

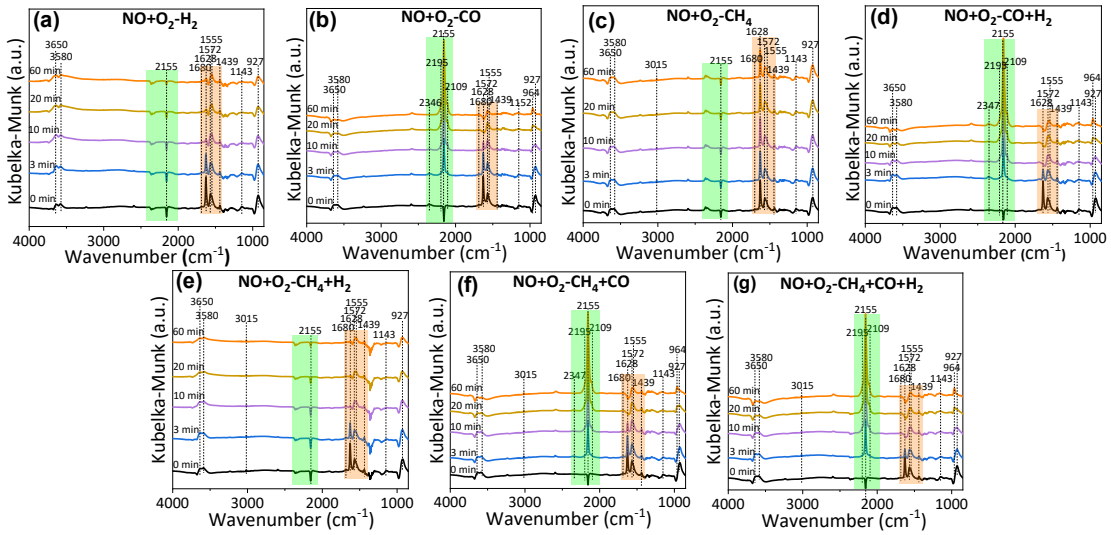

Fig. S4. *In situ* DRIFTS spectra of (a) H_2, (b) CO, (c) CH_4, (d) CO + H_2, (e) CH_4 + H_2, (f) CH_4 + CO, and (g) CH_4 + CO + H_2 reacted with pre-adsorbed NO + O_2 on Cu-ZSM-5 catalyst at 250 °C as a function of time.

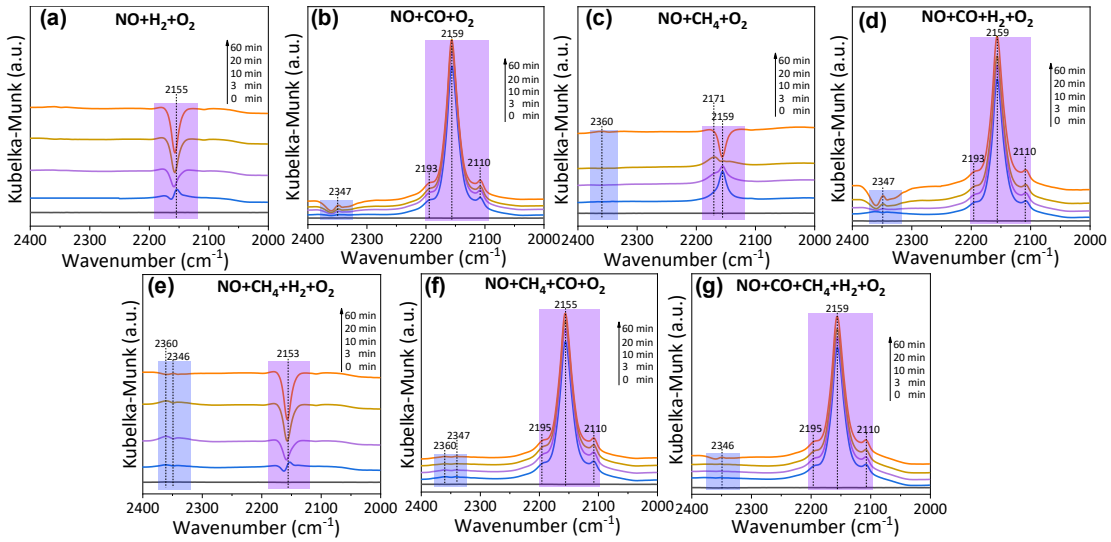

Fig. S5. *In situ* DRIFTS spectra of (a) NO + H_2 + O_2, (b) NO + CO + O_2, (c) NO + CH_4 + O_2, (d) NO + CO + H_2 + O_2, (e) NO + CH_4 + H_2 + O_2, (f) NO + CH_4 + CO + O_2 and (g) NO + CH_4 + CO + H_2 + O_2 on Cu-ZSM-5 catalyst at 250 °C as a function of time.

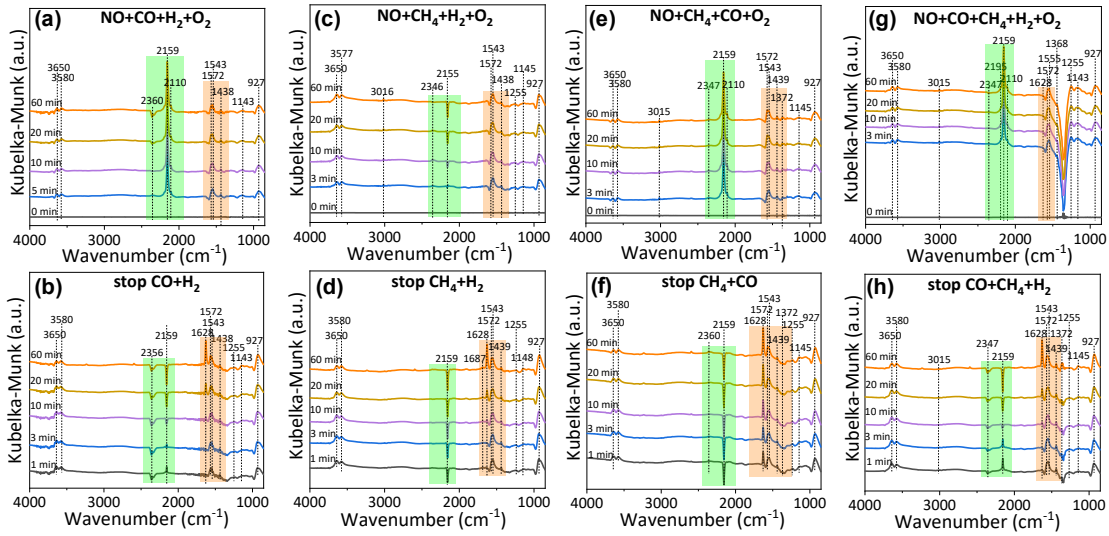

Fig. S6. *In situ* DRIFTS spectra of (a) NO + CO + H_2 + O_2, (b) stop CO + H_2, (c) NO + CH_4 + H_2 + O_2, (d) stop CH_4 + H_2, (e) NO + CH_4 + CO + O_2, (f) stop CH_4 + CO (e) NO + CH_4 + CO + H_2 + O_2 and (f) stop CH_4 + CO + H_2 on Cu-ZSM-5 catalyst at 250 °C as a function of time.

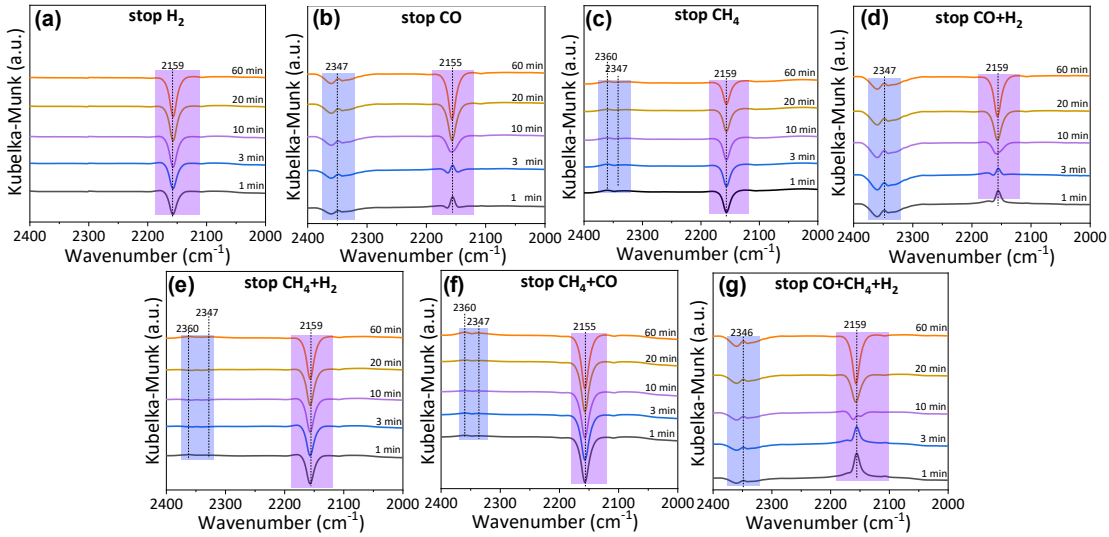

Fig. S7. *In situ* DRIFTS spectra of (a) stop H_2, (b) stop CO, (c) stop CH_4, (d) stop CO + H_2, (e) stop CH_4 + H_2, (f) stop CH_4 + CO and (g) stop CH_4 + CO + H_2 on Cu-ZSM-5 catalyst at 250 °C as a function of time.

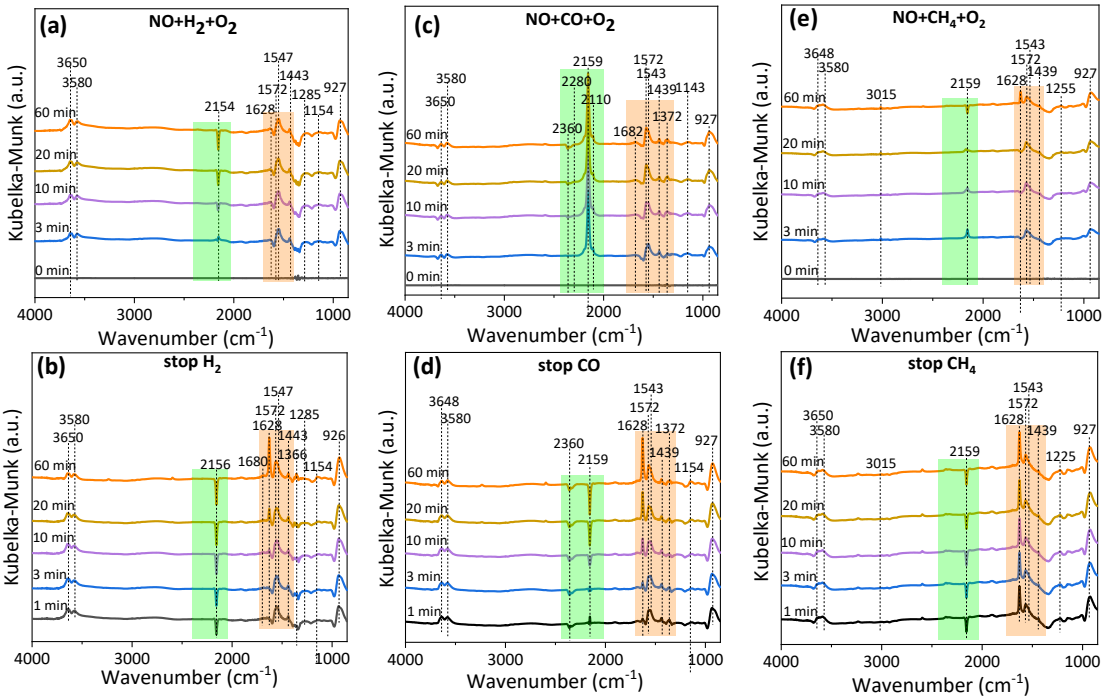

Fig. S8. *In situ* DRIFTS spectra of (a) NO + H_2 + O_2, (b) stop H_2, (c) NO + CO + O_2, (d) stop CO, (e) NO + CH_4 + O_2 and (f) stop CH_4 on Cu-ZSM-5 catalyst at 250 °C as a function of time.

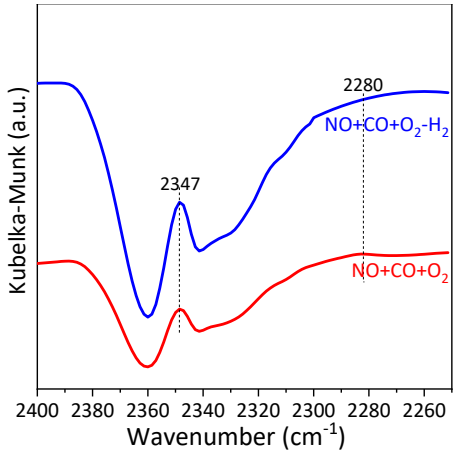

Fig. S9. *In situ* DRIFTS spectra on Cu-ZSM-5 catalyst at 250 °C under different reaction conditions.

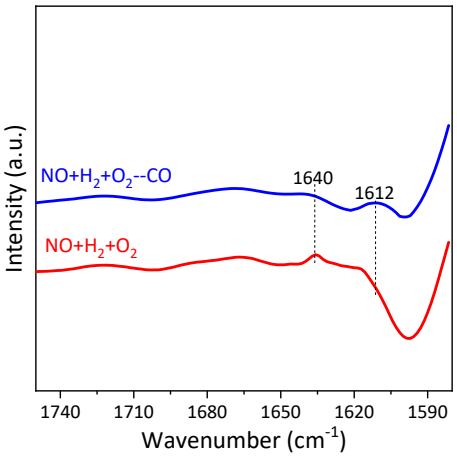

Fig. S10. *In situ* DRIFTS spectra on Cu-ZSM-5 catalyst at 350 °C under different reaction conditions.

Table S1. Observed species in the reaction process as identified by IR.

Wavenumber (cm^{-1})	Species and mode	Ref.
1143	Adsorbed NO	4
1255	Bridged nitrates	4
1439	Nitrites	5
2155	NO^+ species	6
964	Bidentate carbonates	7
1372	Monodentate carbonate	8
2109/2155	$Cu^+(CO)$ carbonyl	9, 10
2195	$Cu^+(CO)_2$ carbonyl	11
2346/2360	Adsorbed CO_2	12
1285	Coordinated NH_3	13
3015	Adsorbed CH_4	14
3580	Si-OH-Al	15
3650	Cu-OH	15

Table S2. Reaction steps of various SCR reactions.

Reaction type	Reaction steps	No.
NO adsorption and nitrate formation	$NO + * \rightarrow NO*$	R1
	$O_2 + * \rightarrow O_2*$	R2
	$NO* + *\rightarrow N* + O*$	R3
	$O_2* + *\rightarrow O* + O*$	R4
	$N* + N* \rightarrow N_2 + 2*$	R5
	$NO* + N* \rightarrow N_2O + 2*$	R6
	$NO* + O_2* \rightarrow NO_2* + O*$	R7
	$NO* + O_2{}^{-}* \rightarrow NO_3{}^{-}* + *$	R8
	$N_2O* \rightarrow N_2 + O*$	R9
H_2-SCR	$H_2 + * \rightarrow H_2*$	Ra1
	$H_2* \rightarrow 2H*$	Ra2
	$N* + H* \rightarrow NH* + *$	Ra3
	$NH* + H* \rightarrow NH_2* + *$	Ra4
	$NH_2* + H* \rightarrow NH_3* + *$	Ra5
	$O* + H* \rightarrow OH* + *$	Ra6
	$OH* + H* \rightarrow H_2O + 2*$	Ra7
	$NO_2* + H_2* \rightarrow ONH* + OH*$	Ra8
	$ONH* + NO* + H* \rightarrow N_2 + H_2O + O* + 2*$	Ra9
	$ONH* + NO* + H* \rightarrow N_2O* + H_2O + 2*$	Ra10
	$4NH_3 + 4NO + 2O_2 \rightarrow 4N_2 + 6H_2O$	Ra11
	$2NH_3 + NO + NO_2 \rightarrow 2N_2 + 3H_2O$	Ra12
CO-SCR	$CO + * \rightarrow CO*$	Rb1
	$N* + CO* \rightarrow NCO* +*$	Rb2
	$NO_2* + CO* \rightarrow NCO* + O_2*$	Rb3
	$CO* + 2NO_2* \rightarrow N_2 + CO_2 + 3O*$	Rb4
	$NCO* + NO* \rightarrow N_2 + CO_2 + 2*$	Rb5
	$CO* + O* \rightarrow CO_2 + 2*$	Rb6
	$2NCO* + 3H_2O \rightarrow 2NH_3 + 2CO_2 + O*$	Rb7
	$NCO* + NO_2* \rightarrow N_2 + CO_2 + * + O*$	Rb8
CH_4-SCR	$CH_4* + O* \rightarrow CH_3* + OH*$	Rc1
	$CH_3* + O* \rightarrow CH_2* + OH*$	Rc2
	$NO* + CH_3* \rightarrow CH_3NO* + *$	Rc3
	$4CH_3NO + 2NO + 4O_2 \rightarrow 3N_2 + 6H_2O + 4CO_2$	Rc4
	$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$	Rc5
	$2NO + CH_4+ O_2 \rightarrow N_2 + CO_2 + 2H_2O$	Rc6