Efficient and stable catalyst of α-FeOOH for NO oxidation from coke oven flue gas by the catalytic decomposition of gaseous H_2O_2

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Experimental Section

NO temperature-programmed desorption (NO-TPD) was conducted on a fixed-bed reactor equipped with a Fourier transform infrared (FTIR) spectrometer (gas cell). 0.1 g of sample was pretreated in N_2 at 120 °C for 0.5 h. After cooled to 30 °C, the sample was exposed to 0.5 vol% NO/N_2 for 1 h, followed by a N_2 purge at 50 °C for 0.5 h to remove physisorbed NO. Finally, NO-TPD was measured from 50 °C to 350 °C.

The experimental equipment for evaluating catalytic performance of α-FeOOH is shown in **Fig. S1**. It includes a simulated flue gas generation system, a NO catalytic oxidation system, and a flue gas analysis system. The dosage of catalyst used in this process was 0.5 g. The concentrations of NO, SO_2 and O_2 in simulated flue gas were 200 ppm (parts per million by volume), 660 ppm and 6.0%, respectively. The total flow rate of simulated flue gas was kept at 4.0 L/min in all experiments. The gaseous H_2O_2

was generated by an evaporative heater, and carried to simulated flue gas by N_2. H_2O_2 solution was carried to the evaporative heater via a peristaltic pump. The vaporized temperature was controlled at 120 °C to avoid H_2O_2 decomposition. To avoid the water vapour condensing, the pipeline was heated at about 120 °C. The vaporized H_2O_2 was mixed with simulated flue gas, and then the mixture gas contacted with the catalyst, in which gaseous H_2O_2 decomposed into radicals over the catalyst and oxidized NO into NO_2 or HNO_3. The tail gas was absorbed by a $KMnO_4/H_2SO_4$ solution.

The conversion of NO_2 and SO_2 was calculated as follows:

$$Conversion = \frac{C_{in} - C_{out}}{C_{in}} \times 100\%,$$

where C_{in} and C_{out} are the concentrations of NO or SO_2 at the inlet and outlet of the reactor, respectively.

$$NO_2\, yield = \frac{C_{NO_2,out}}{C_{NO,in}} \times 100\%,$$

where $C_{\mathrm{NO,\ in}}$ is NO concentration at the inlet and $C_{NO_2,out}$ is NO_2 concentration at the outlet of the catalyst reactor.

The concentrations of NO, NO_2, and SO_2 are recorded after one experiment is conducted for 30 min.

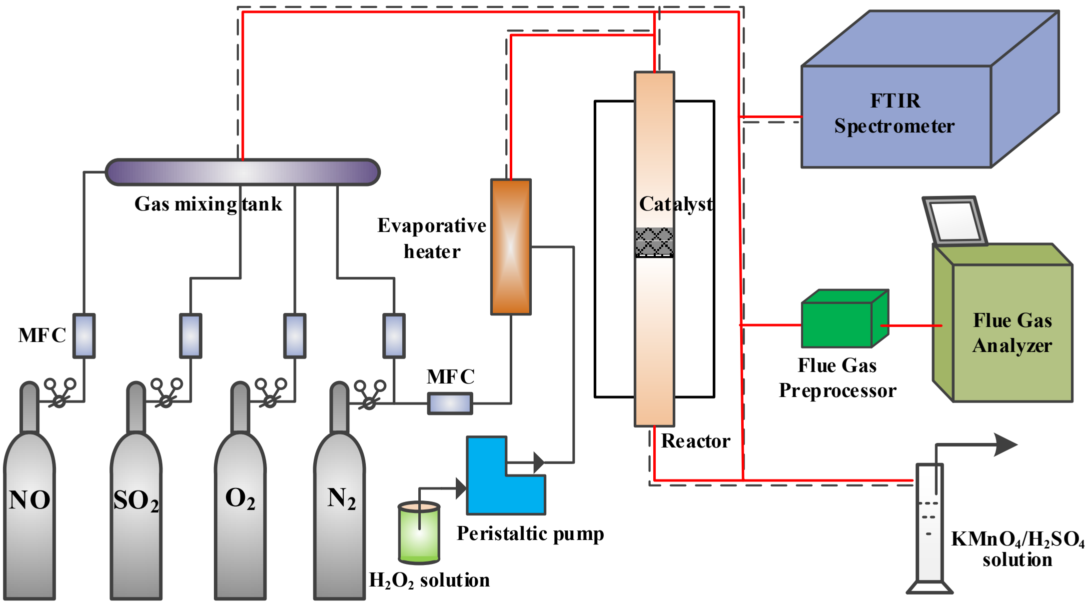


Fig. S1 The experimental equipment for evaluating the catalytic performance of α-FeOOH.

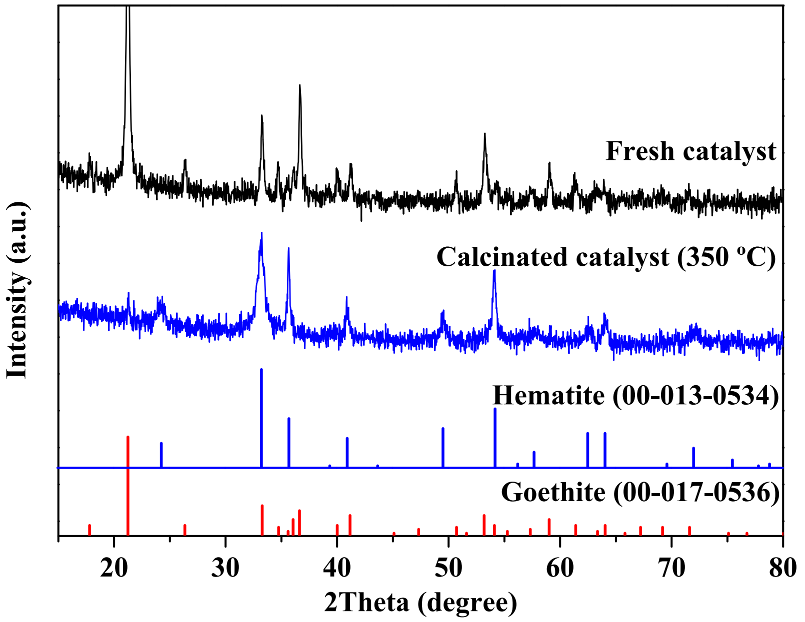


Fig. S2 XRD spectra of the fresh catalyst and the catalyst calcinated at 350 °C.

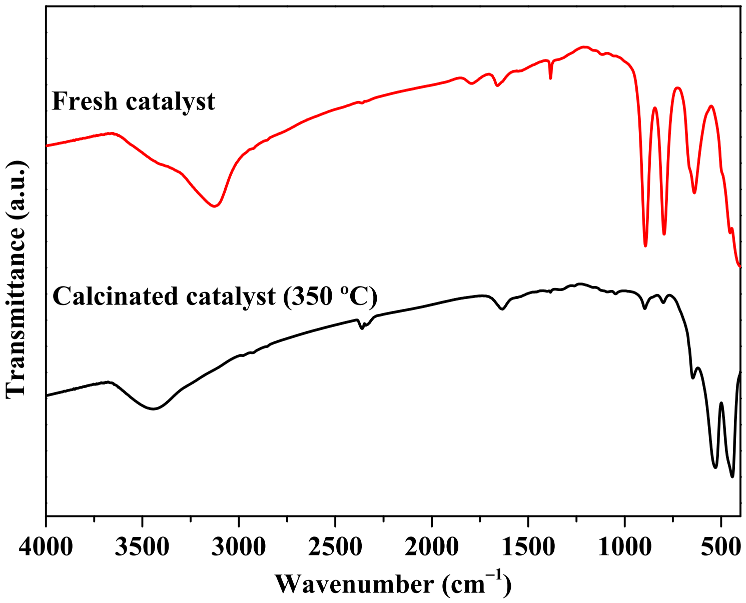
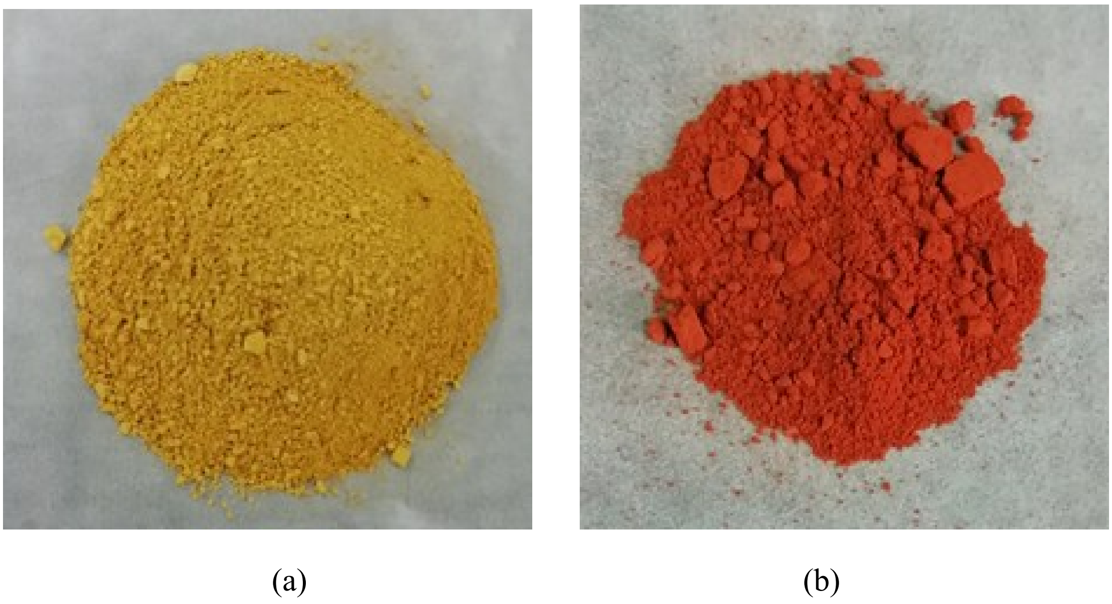


Fig. S3 FTIR spectra of the fresh catalyst and the catalyst calcinated at 350 °C.


(a)

(b)

Fig. S4 Images of the fresh catalyst (a) and the catalyst calcinated at 350 °C (b).

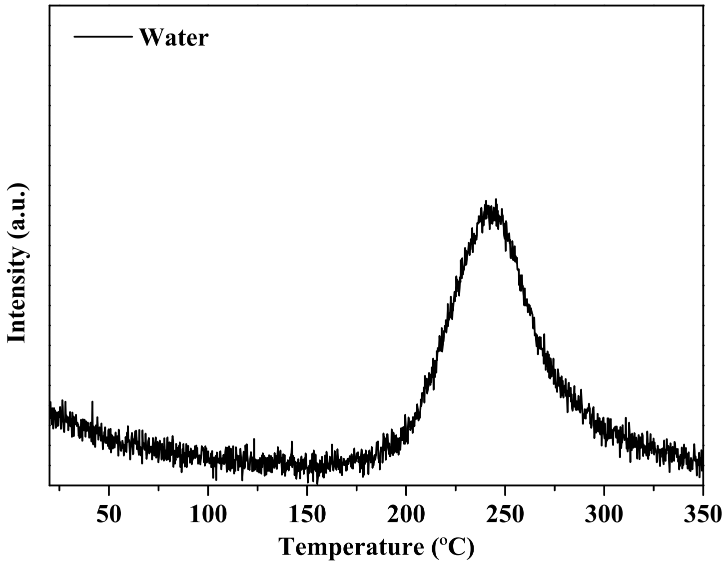


Fig. S5 The decomposition of α-FeOOH.

Table S1. A comparison of simultaneous removal of NOx and SO_2 based on the catalytic decomposition of H_2O_2 over catalysts.

Catalysts	H_2O_2/NO	The amount of catalyst vs gas flow rate (g/(mL/min))	GHSV (h^{-1})	NO removal efficiency (%)	Stability	References
Fe_2O_3	≈660	0.6/240	–	80	-	J. Ding (2014)[1]
$Fe_2(SO_4)_3$	2.5	2/1500	–	92.5	Reducing by 17.5% (12 h)	B. Wu (2018)[2]
nZVI (Fe^0)	≈22	0.3/2600	198,726	80.4	Reducing by 7.6% (5 h)	Y. Zhao (2017)[3]
Modified fly ash	3.0	2.0/300	–	80	Reducing by 3.0% (16 h)	B. Yang (2019)[4]
$Fe_2(MoO_4)_3$	1.5	2.0/1500	84,758	91.4	Reducing by 4.1% (15 h)	X. Liu (2019)[5]
α-FeOOH	2.0	0.5/4000	137,747	98.8 (NO conversion)	Reducing by ~8.8% (45 h)	This work

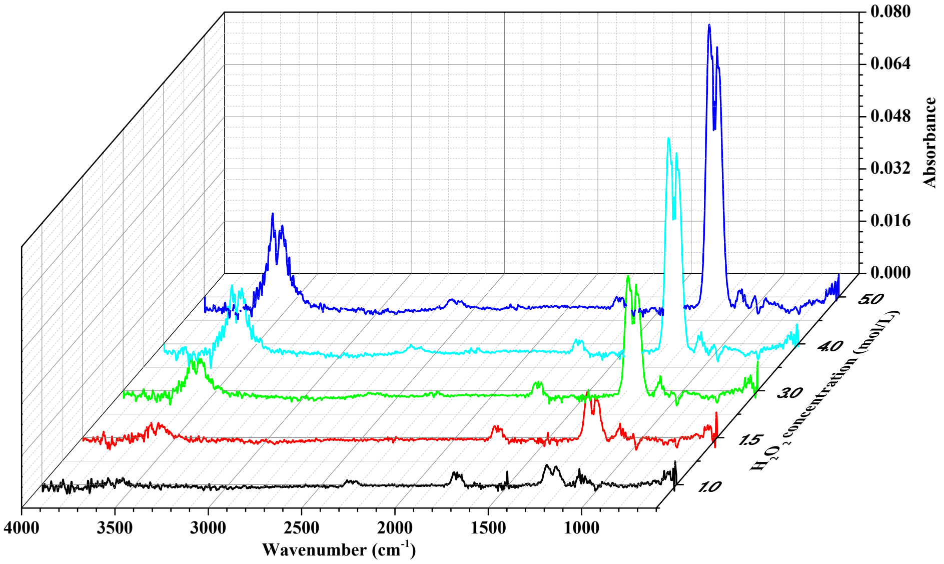


Fig. S6 FTIR spectra of gaseous H_2O_2 under the different H_2O_2 concentration.

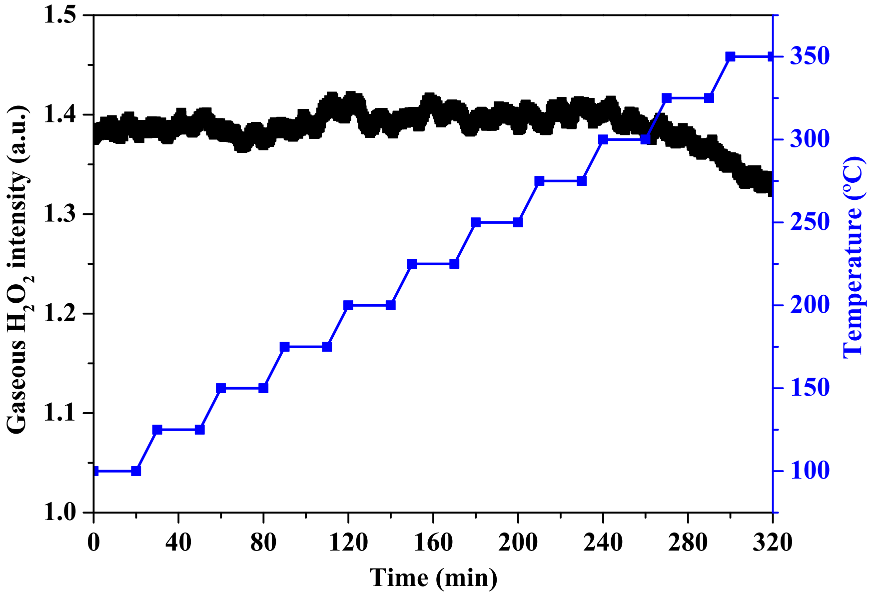


Fig. S7 The decomposition of gaseous H_2O_2 under different temperature.

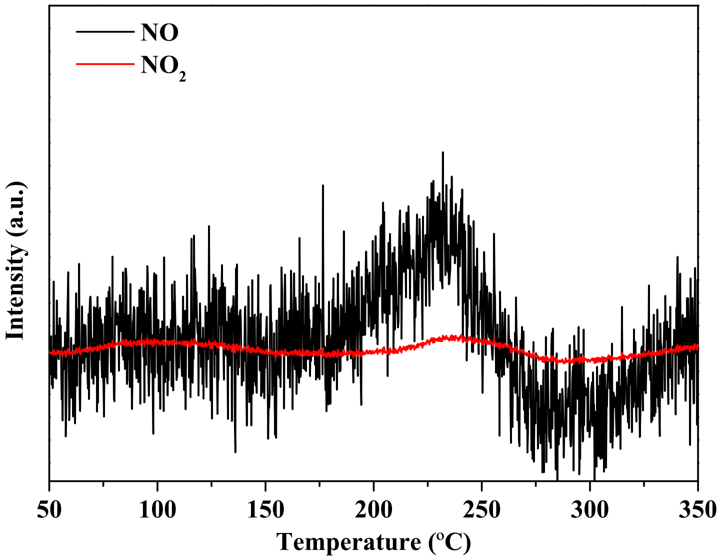


Fig. S8 NO-TPD of α-FeOOH.

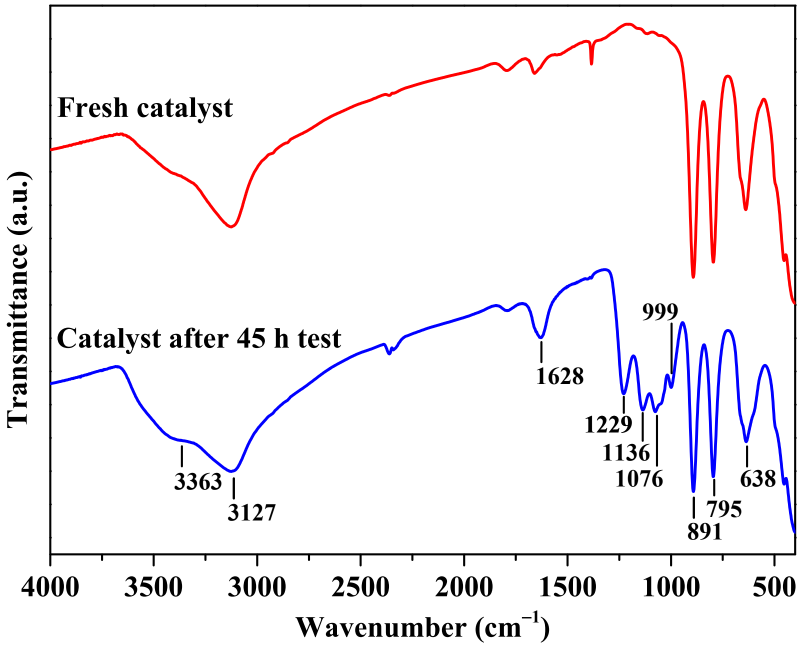


Fig. S9 FTIR spectra of the fresh catalyst and the used catalyst (45 h test).

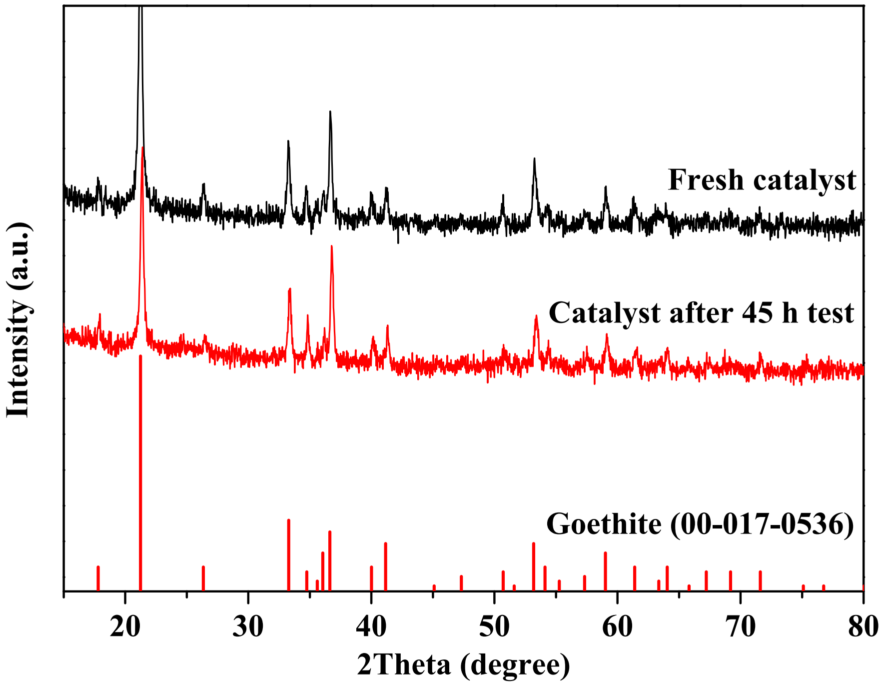


Fig. S10 XRD spectra of the fresh catalyst and the used catalyst (45 h test).

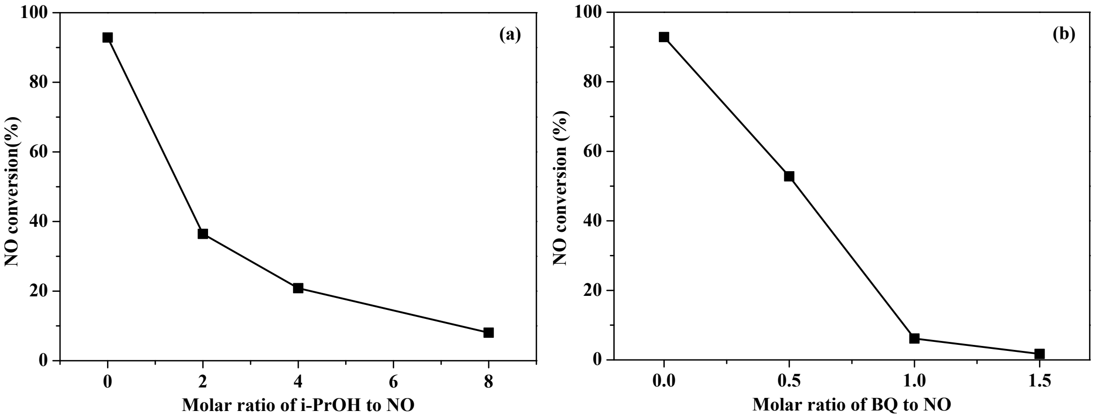


Fig. S11 Effects of the molar ratio of (a) i-PrOH and (b) BQ to H_2O_2 on NO conversion. (Temperature, 225 °C; other experimental parameters were the same as that in **Fig. 1**)

References

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