Removal mechanism and quantitative control of trichloroethylene in post-plasma-catalytic system over Mn-Ce/HZSM-5 catalysts

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Text S1 The calculation method of TCE removal efficiency, CO_2 yield and CO yield

The TCE removal efficiency (η_{TCE}), CO_2 yield (Y_{CO_2}) and CO yield (Y_{CO}) were calculated as follows:

$$\eta_{TCE}(\%)=\frac{[TCE]_{inlet}-[TCE]_{outlet}}{[TCE]_{inlet}}\times 100\% \quad (1)$$

$$Y_{CO_2}(\%)=\frac{[CO_2]}{2[TCE]_{inlet}}\times 100\% \quad (2)$$

$$Y_{CO}(\%)=\frac{[CO]}{2[TCE]_{inlet}}\times 100\% \quad (3)$$

where $[TCE]_{inlet}$ and $[TCE]_{outlet}$ are denoted as the TCE concentrations at the inlet and outlet of the reactor, respectively; $[CO_2]$ and $[CO]$ are represented as the CO_2 and CO concentrations at the exit of the reactor, respectively.

Text S2 Electrical measurement method

The reactor was connected to a DC high voltage power supply (SR40-R-1200, Technix). A high voltage probe (Fluke 80 K-40, division ratio 1/1000) was used to measure the applied voltage. The discharge current was determined by monitoring the voltage signal across a 100 Ω resistor which was put in series between the counter electrode and ground. The discharge power (P) was calculated using the applied voltage (U) and the reactor current (I) according to the Eq. (4).

$$P\ (\mathrm{W})=U\times I \quad (4)$$

Text S3 Catalyst preparation and characterization

The Mn-Ce/HZSM-5 (molar ratio of Ce/Mn=0.2, 0.6, 1 and 1.4) catalysts were prepared using the deposition–precipitation method. A typical synthesis of Mn-Ce/HZSM-5 contained the following steps. The $Ce(NO_3)_3\cdot 6H_2O$ and $Mn(NO_3)_2$ were dissolved in deionized water with continuous stirring. Then, HZSM-5, KMnO4 and Na_2CO_3 were added to the above mixture successively under stirring. Subsequently,

the obtained mixture aged for 4 h at 60 ℃. Finally, the above mixture was filtered, dried overnight at 70 ℃, followed by calcination in air at 500 ℃ for 3 h. The catalyst samples were donated as $MnCe_x$, where x stood for the molar ratio of Ce/Mn.

The XRD patterns of all the prepared samples were analyzed by a X-ray powder diffractometer (PAN analytical, X'pert, Almelo) equipped with a Cu-Kα radiation source (λ = 1.5406 Å). The Raman spectra were acquired on Confocal Raman spectroscopy (Lab RAM, HR 800, Horiba, France) using the 514.0 nm radiation from an argon laser. The quantitative analysis of the element content of all the prepared samples used an inductively coupled plasma emission spectrometry (E 9000, Shimadzu). The specific surface area, total pore volume and average pore diameter of all the prepared samples were obtained via N_2 adsorption–desorption isotherms at 77 K using an SSA-4200 analyzer (Beijing Builder). The morphology of all the prepared samples was recorded through field-emission scanning electron microscopy (SEM 500, Gemini). The high-resolution transmission electron microscopy (HR-TEM) was used to measure the dispersion and configuration of catalysts (JEOL, Model JEM-2100HR instrument, Japan). H_2-TPR and NH_3-TPD of all the prepared samples were both operated on the same Micromeritics AutoChem 2910 instrument equipped with a thermal conductivity detector (TCD), with 0.05 g catalyst for H_2-TPR and 0.1 g catalyst for NH_3-TPD. XPS was conducted at room temperature on AXIS ULtrabld XPS (ESCALAB Xi+, Thermo Fisher Scientific) equipment to analyze the oxidation states of the elements. The C1s photoelectron peak at 284.8 eV was used to check the binding energy (BE) calibration.

Text S4 The second-order polynomial used to fit the modelling response variable

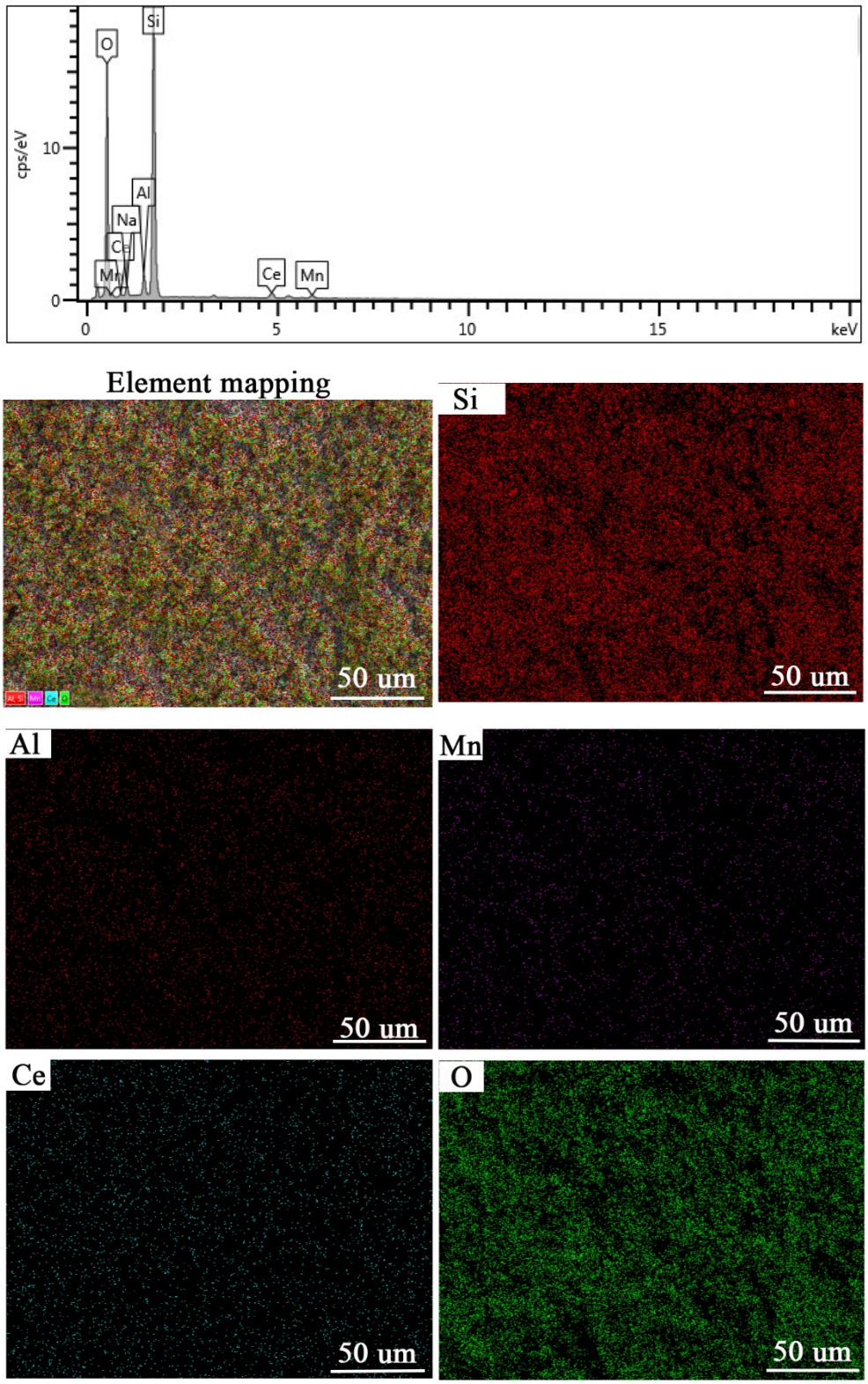
$$Y = \beta_0 + \sum_{i=1}^{k} \beta_i X_i + \sum_{i=1}^{k} \beta_{ii} X_i^2 + \sum_{i=1}^{k-1} \sum_{j=2}^{k} \beta_{ij} X_i X_j + \varepsilon \quad (5)$$

Y: the predicted response; k: the number of variables, ε: the residual value: β_0: the constant; β_i: the linear coefficient; β_{ii}: the quadratic coefficient; β_{ij}: the interaction coefficient; X_i, X_j: the coded independent variables.

Figure Captions

Fig. S1. EDX mapping of $MnCe_1$ sample.

Fig. S2. Predicted and actual results of (a) TCE removal efficiency, (b) CO_2 yield and (c) CO yield.



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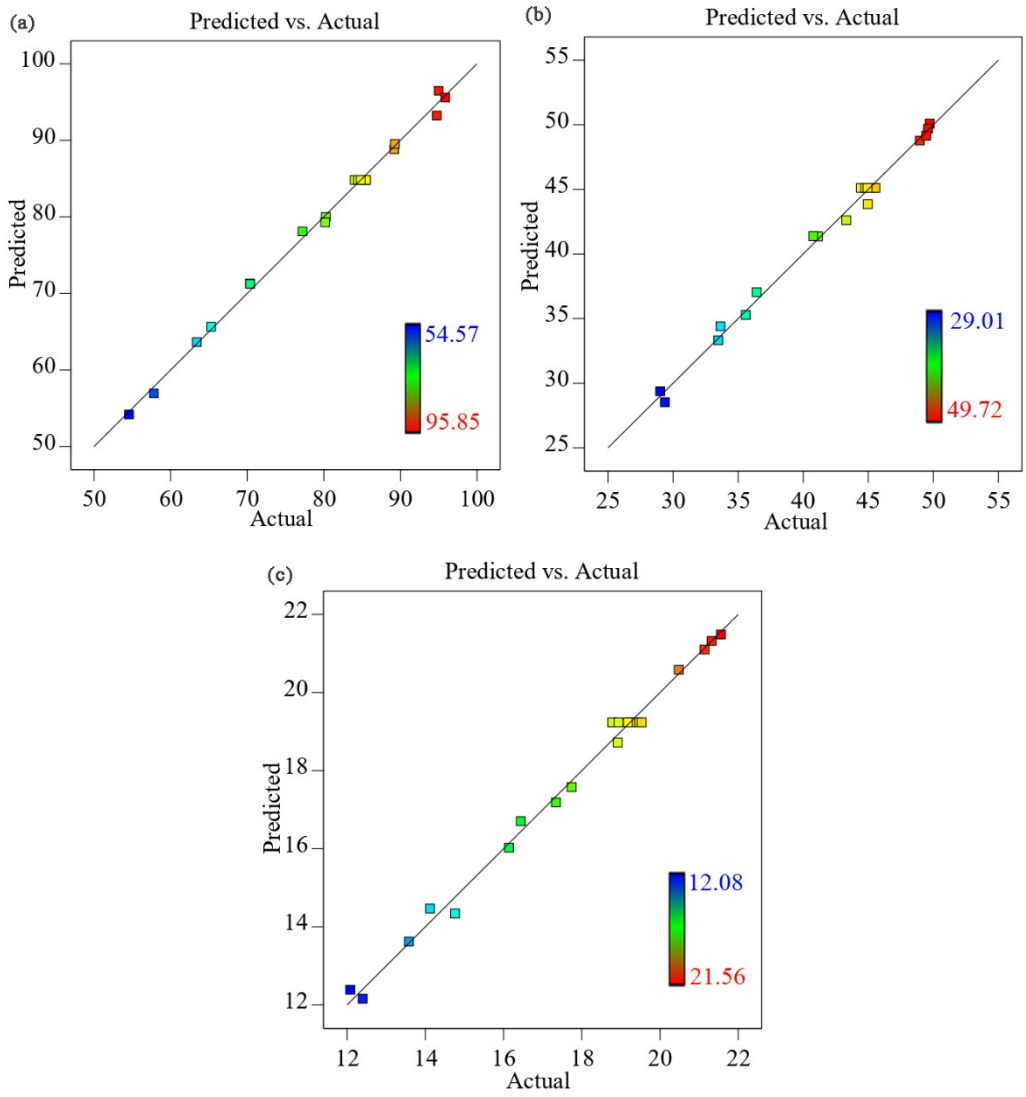

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