Electronic Supplementary Information

Vanadium tetrasulfide cross-linking graphene-like carbon driving sustainable electron supply chain of pollutants through activation of dissolved oxygen and hydrogen peroxide

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Fig. S1.The structure of the used organic pollutants.

Detailed process and analytical methods

Pollutants concentration test methods. Dye contaminants were analyzed by UV spectrophotometer (752 N) with wavelength settings of 554, 669 and 485 nm for RhB, MB, AO7, respectively. Other colorless pollutants were analyzed using a 1200 series HPLC (Agilent, U.S.A.) equipped with a UV detector and a ZORBAX Eclipse XDB- C_{18} column (4.6×150 mm, 5 µm). During the test, the mobile phase of BPA is methanol/ultrapure water=70/30, and the wavelength is set to 225 nm; the mobile phase of 2-CP is methanol/ultrapure water=60/40, and the wavelength is set to 275 nm; the mobile phase of CIP is acetonitrile/water solution containing 0.08% phosphoric acid=70/30, and the wavelength is set to 278 nm.

HPLC-MS analysis was conducted in auto full-scan mode (Agilent 1260-6460). The samples were prepared according to the following steps: 0.6 g L^{-1} catalyst was added to 10 mg L⁻¹ aqueous RhB solution, the suspension was stirred for 5 min at room temperature, 1 mL of the suspension was collected and filtered, and the samples were tested in HPLS-MS with the mobile phase set at methanol/ultrapure water=60/40 and the flow rate set at 0.2 mL/min.

Detection of 'OH and HO₂'/O₂⁻⁻ EPR signals.^{1, 2} BMPO-trapped EPR signals were detected in different air-saturated methanol/aqueous dispersions of the corresponding samples using a Bruker A300-10/12 EPR spectrometer at room temperature.

To detect **'OH**: Typically, in the absence of H_2O_2 , add 0.005 g of the prepared powder sample to 500 μ L of water or RhB (100 mg L⁻¹, prepare with deionized water), and shake the mixture well for 2 min. Then, add 100 μ L mixture to the centrifuge tube

containing 20 μ L BMPO (250 mM), thoroughly incorporate, let the solution stand for 1 min, and draw it into a capillary for detection. In the presence of H₂O₂, add 0.005 g of the prepared powder sample and 100 μ L H₂O₂ to 500 μ L of water or RhB (100 mg L⁻¹, prepare with deionized water). Then, add 100 μ L mixture to the centrifuge tube containing 20 μ L BMPO (250 mM), thoroughly incorporate, let the solution stand for 1 min, and draw it into a capillary for detection.

To detect HO_2'/O_2 in the absence of H_2O_2 , 0.005 g of the prepared powder sample was added to 500 µL of methanol or RhB (100 mg L⁻¹, prepare with methanol). In the presence of H_2O_2 , add 0.005 g of the prepared powder sample and 100 µL H_2O_2 to 500 µL of methanol or RhB (100 mg L⁻¹, prepare with methanol). Then, perform other determination steps as above.

In situ Raman analysis was performed to reflect the actual reaction process of the catalyst, BPA and H_2O_2 .³ First, H_2O_2 was added to H_2O/BPA solution to determine the location of the H_2O_2 peak. VSO-C(π) was also added to the H_2O/BPA solution for analysis. Then, the aqueous samples were collected from the actual reaction suspensions at 1 min, 4 min, 8 min and 15 min after H_2O_2 and catalyst were added. The collected suspension was placed into the slide and scanned from 100–4000 cm⁻¹ using an XploRA confocal spectrometer (Jobin Yvon, Horiba Gr, Palaiseau, France) with a 532-nm Nd:YAG laser light source.

CV curve were measured on an electrochemical workstation (Shanghai chenhua instrument co., LTD.) with three standard electrodes. The working electrode was prepared from the sample material coated on conductive glass, the reference electrode was extremely saturated calomel, a platinum electrode was used as the pair electrode,

and the electrolyte was 0.1 M Na₂SO₄ solution or 10 mg L^{-1} RhB prepared with 0.1 M of Na₂SO₄.



Fig. S2. XPS survey spectrum of VSO- $C(\pi)$.



Fig.S3. UV diffuse reflectance spectrum of VSO-C(π) and VSO-C(π) after reaction.



Fig.S4. HPLC-MS spectra of (a) Pre-reaction contaminant solution and (b) (c)

Intermediate products detected in VSO-C(π)/RhB/DO systems after 5 min.



Fig.S5. Effects of different gas filling in the whole process on removal of RhB by VSO-C(π). Reaction conditions: natural initial pH, initial pollutant concentration 10 mg L⁻¹, catalyst of 0.6 g L⁻¹. (gas was injected into the system for 30 min before the reaction, and the gas was kept filled during the reaction.)



Fig. S6. In situ Raman spectra of (a) VSO-C_{(π)/H₂O and (b) VSO-C(π)/pollutant aqueous dispersions recorded at different times after adding H₂O₂.}

Notes and references

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