

Supporting information

Removal of *p*-toluenesulfonic acid from wastewater using a filtration-enhanced electro-Fenton reactor

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Text S1. Analysis of PTSA. The concentration of PTSA was analyzed using reversed-phase HPLC (Agilent 1290, USA) with a C18 column (ZORBAX Eclipse Plus, 250 mm × 4.6 μm, 5 μm). A 50:50 v/v methanol-phosphate buffer (pH 2.9) was used as the mobile phase. The constant flow rate was set at 1.0 mL/min. An Agilent 1200 photodiode array detector (DAD) at 221 nm was used to detect PTSA, with the injection volume of 50 μL and test temperature of 40°C.

To identify the degradation intermediates of PTSA, the samples were purified using solid-phase extraction (SPE) column to remove salinity prior to the measurement by a liquid chromatograph mass spectrometer (LC-MS) (Waters, Thermo, Q-Exactive) equipped with ACQUITY UPLC HSS T3 column (2.1 mm × 100 mm, 1.8 μm). A 0.05% formic-acetonitrile mixture was used as the mobile phase at the flow rate of 0.3 mL/min. The electrospray ionization mass spectrometry was performed at positive and negative modes with a spray voltage of 3.0 kV and 3.2 kV, respectively. The heater temperature was set at 300°C, ions with m/z 20–200 that cover each frequently-detected PTSA intermediates was monitored.

Figures

Fig. S1. The LSV curve of graphite plate anode and stainless-steel mesh cathode.

Fig. S2. The concentration of H₂O₂ in the effluent of the FEEF system under the experimental conditions of [PTSA]₀ = 100 mg/L, voltage = 2.5 V, pH = 3.0, 0.2 mM Fe²⁺ and 1.0 mM H₂O₂ dosage.

Fig. S3. The total ion chromatograms (TIC) for the PTSA effluent of FEEF system at 120 min (a) at positive mode, and (b) negative mode.

Fig. S4. LC-MS spectra for the PTSA intermediate products in the effluent of FEEF system at positive mode.

Fig. S5. LC-MS spectra for the PTSA intermediate products in the effluent of FEEF system at negative mode.

Fig. S6. The removal efficiency of COD in MCEF system under the experimental conditions of $[\text{PTSA}]_0 = 100 \text{ mg/L}$, voltage = 2.5 V, pH = 3.0, 0.2 mM Fe^{2+} and 1.0 mM H_2O_2 dosage.

Tables

Table S1. Treatment cost of pre-unit PTSA removal in three systems

Table S2 Comparison of PTSA removal efficiency with other system literature results.

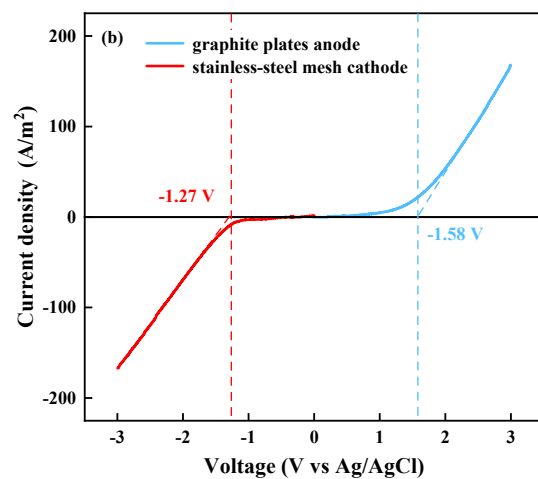


Fig. S1. The LSV curve of graphite plate anode and stainless-steel mesh cathode.

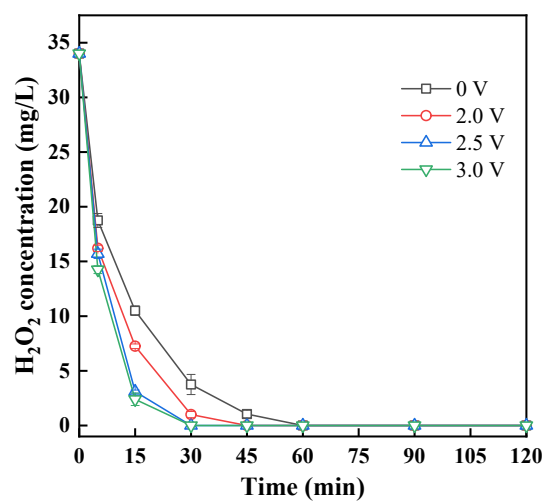


Fig. S2. The concentration of H₂O₂ in the effluent of the FEEF system under the experimental conditions of [PTSA]₀ = 100 mg/L, voltage = 2.5 V, pH = 3.0, 0.2 mM Fe²⁺ and 1.0 mM H₂O₂ dosage.

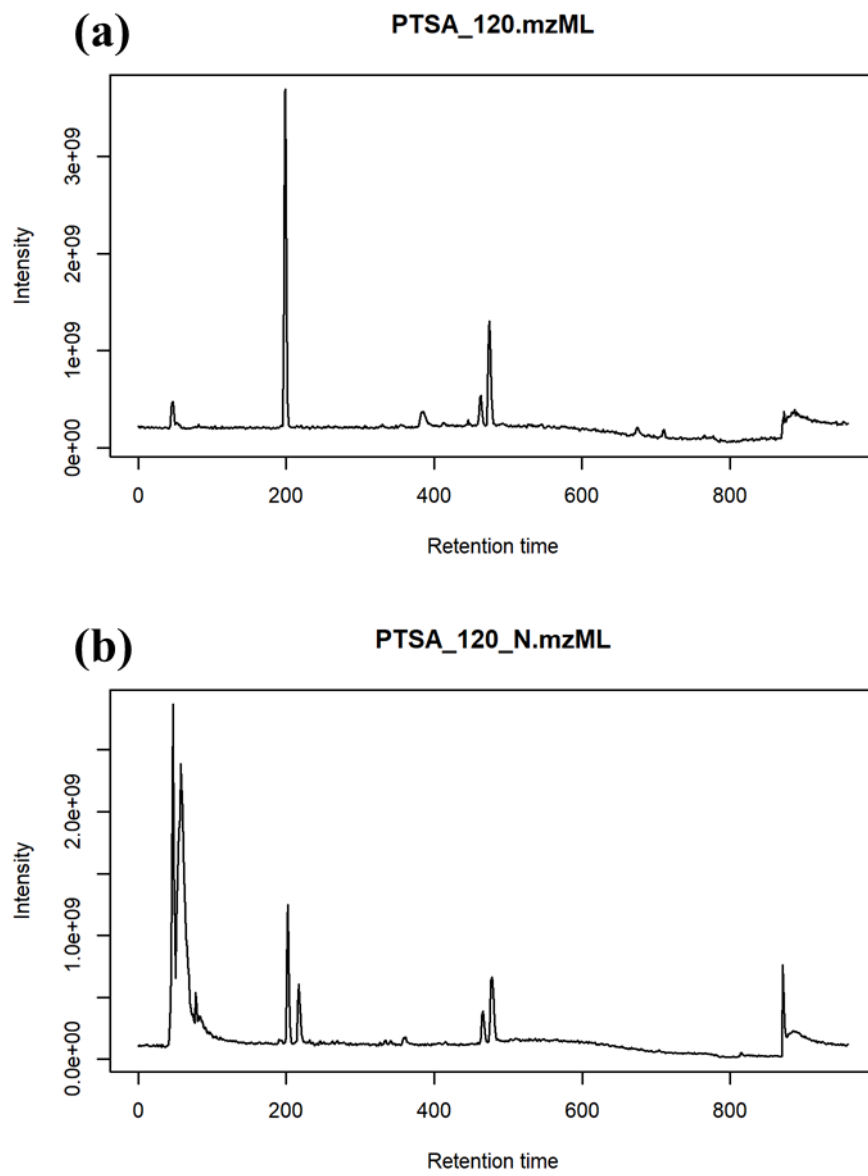


Fig. S3. The total ion chromatograms (TIC) for the PTSA effluent of FEEF system at 120 min (a) at positive mode, and (b) negative mode.

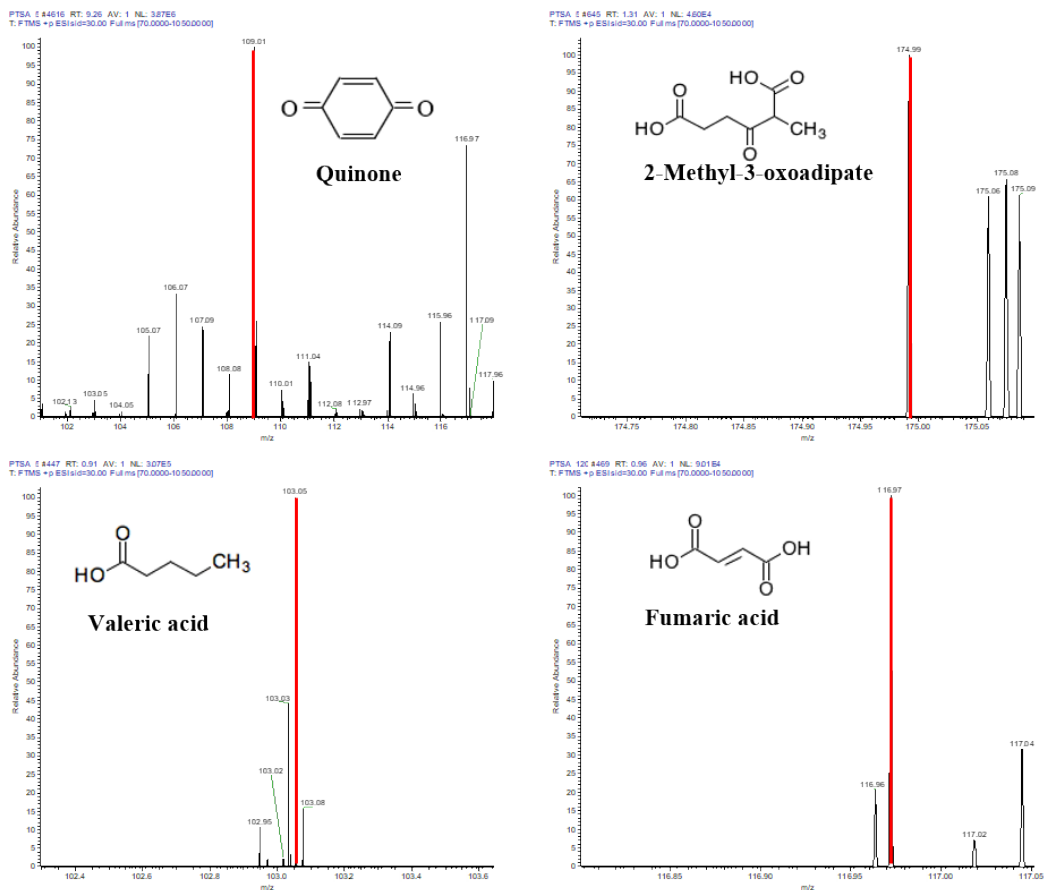


Fig. S4. LC-MS spectra for the PTSA intermediate products in the effluent of FEEF system at positive mode.

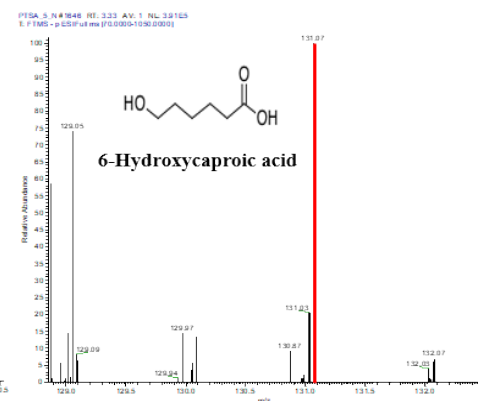
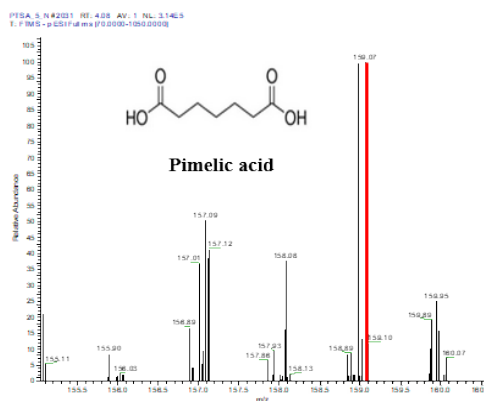
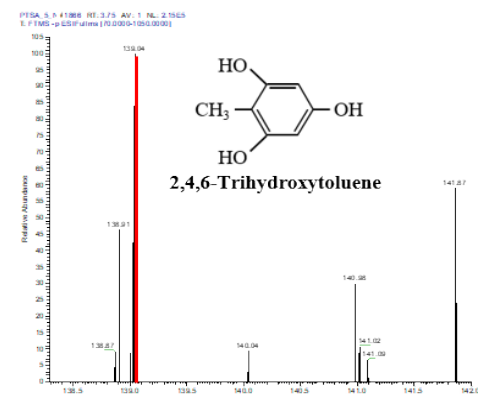
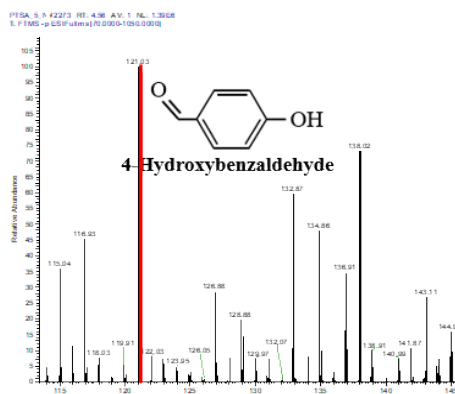
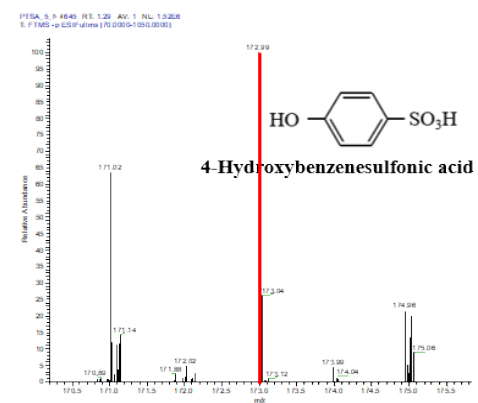
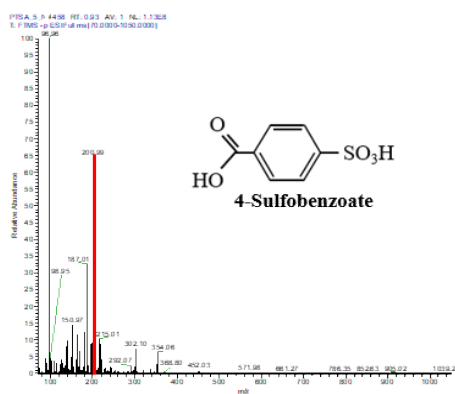
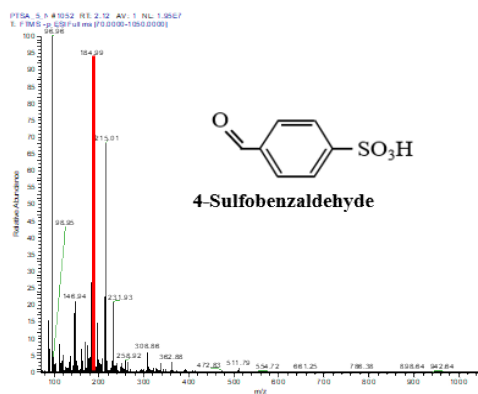
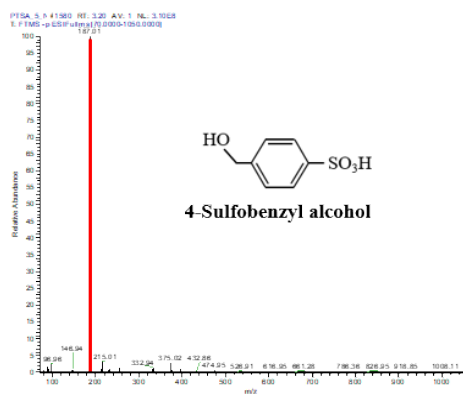


Fig. S5. LC-MS spectra for the PTSA intermediate products in the effluent of FEEF system at negative mode.

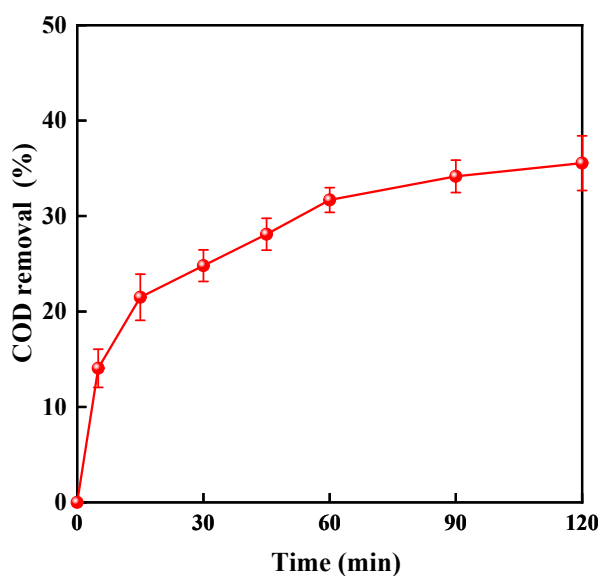


Fig. S6. The removal efficiency of COD in MCEF system under the experimental conditions of $[PTSA]_0 = 100$ mg/L, voltage = 2.5 V, pH = 3.0, 0.2 mM Fe^{2+} and 1.0 mM H_2O_2 dosage

Table S1. Treatment cost of pre-unit PTSA removal in three systems.

Items (\$/kg PTSA)	FEEF	Fenton	Anodic oxidation
DC power supply	3.49×10^{-1}	0	3.53
Aeration	4.48×10^{-4}	1.05×10^{-3}	5.23×10^{-3}
98% $FeSO_4 \cdot 7H_2O$	6.06×10^{-2}	1.27×10^{-1}	0
27.5% H_2O_2	3.97×10^{-1}	8.32×10^{-1}	0
iron sludge treatment ^a	5.72×10^{-2}	1.20×10^{-1}	0
Total Cost	8.64×10^{-1}	1.08	3.53

^a The price of iron sludge disposal is obtained from a solid waste disposal enterprise (Shanghai Environment Group Co., Ltd.) in China.

Table S2 Comparison of PTSA removal efficiency with other system literature results.

Process	Reaction conditions	The initial Concentration of ammonia	Removal efficiency	Reference
Catalytic ozonation (Co _x Fe ₁ LDH)	Catalyst dosage 0.1 g/L, aqueous ozone concentration 6.2 mg/L	20 mg/L PTSA	85% PTSA removal	1
Combined zero- valent iron (ZVI)- photo-Fenton process	Commercial steel wool, H ₂ O ₂ 24 mg/L, UV 300 nm	5 mg/L PTSA	60% PTSA removal	2
Photo-Fenton	Iron (III) chloride 20 mg/L, H ₂ O ₂ 1.0 mM	861 mg/L PTSA	6 h, 47% PTSA removal	3
Filtration-enhanced electro-Fenton	Applied voltage 2.5 V, Fe ²⁺ 0.2 mM, H ₂ O ₂ 1.0 mM	100 mg/L PTSA	2 h, 98% PTSA removal	This work

Reference

- 1 X. Fu, Y. Huang, Y. Wang, M. Liang, Y. Yang, Z. Jin, J. Yang, S. Hu and L. Li, Ozonation Catalyzed by Co_xFe₁ Layered Double Hydroxide for the Degradation of *P*-toluenesulfonic Acid, *Ozone Sci. Eng.*, 2021, **43**, 163–172.
- 2 L. Santos-Juanes, F. S. García Einschlag, A. M. Amat and A. Arques, Combining ZVI reduction with photo-Fenton process for the removal of persistent pollutants, *Chem. Eng. J.*, 2017, **310**, 484–490.
- 3 A. M. Amat, A. Arques, F. López and M. A. Miranda, Solar photo-catalysis to remove paper mill wastewater pollutants, *Sol. Energy*, 2005, **79**, 393–401.