Supporting Materials

Operations of the PSPC, EF, and PC

The optimal current density of H_2O_2 production was determined in the EF reactor without Fe^{2+} addition within 120 min. The current density of 5 mA/cm^2 resulted in the lowest energy consumption of 29 ± 4 kWh/kg H_2O_2 and the highest average current efficiency of 33.6% ± 3.9% among the different applied current densities (Table S1). The optimal current density of Fe^{2+} production was determined in the PC reactor. To avoid excess iron production in the reactor, low current densities (0, 0.5, 1, and 4 mA/cm^2) were tested for Fe^{2+} production in the anode chamber within 120 min. Among the current densities, 0.5 mA/cm^2 resulted in the highest ratio of Fe^{2+} to total iron molar concentration ([Fe^{2+}]/[total Fe]) of 98.9% ± 1.5%. Therefore, the optimum current densities of 5.0 mA/m^2 for H_2O_2 production and 0.5 mA/m^2 for Fe^{2+} production were selected for the following tests.

In the PSPC, the current density was kept at 5.0 mA/cm^2 (with the applied voltage of 3.33 ± 0.09 V) for 1 s for H_2O_2 production and then 0.0 mA/cm^2 (i.e., open circuit) for 0.3 s. The current density was kept at 0.5 mA/cm^2 (with the applied voltage of 0.33 ± 0.05 V) for 0.3 s for Fe^{2+} production and then of 0.0 mA/cm^2 for 1.0 s (i.e., a pulsed switching frequency of 1:0.3 s). In the EF, the current density of 5.0 mA/cm^2 was used for H_2O_2 production. The totally iron released in the PSPC was determined using the phenanthroline spectrophotometric method (Baba et al., 2015). Under the optimal operation conditions, that is, a pulsed switching frequency of 1:0.3 s, 0.5 mA/cm^2 for Fe^{2+} production, electrolyte volume of 35 mL and running time of 240 min, the totally iron released in the PSPC was 196 ± 20 mg/L. Therefore, 196 ± 20 mg/L Fe^{2+} (calculated by $FeSO_4 \cdot 7H_2O$) similar to the total iron released in the PSPC was added in the sample at the beginning of the EF test. In the PC, the current density of 5.0 mA/cm^2 was used in the reactor ($U_H = U_F = 1.11 \pm 0.13$ V).

Table S1 Average current efficiency and energy consumption of H2O2 production within 120 min

Current density (mA/cm^2)	Energy consumption (kWh/kg H_2O_2)	Average current efficiency (%)
1	37 ± 1	10.1 ± 0.0
5	29 ± 4	33.6 ± 3.9
10	80 ± 12	33.1 ± 4.5
20	168 ± 25	28.6 ± 4.1
30	259 ± 36	16.3 ± 2.4
40	472 ± 14	12.7 ± 1.8
50	718 ± 13	8.0 ± 0.2

References

Baba Y, Yatagai T, Harada T, Kawase Y (2015). Hydroxyl radical generation in the photo-Fenton process: Effects of carboxylic acids on iron redox cycling. Chemical Engineering Journal, 277(Supplement C): 229–241

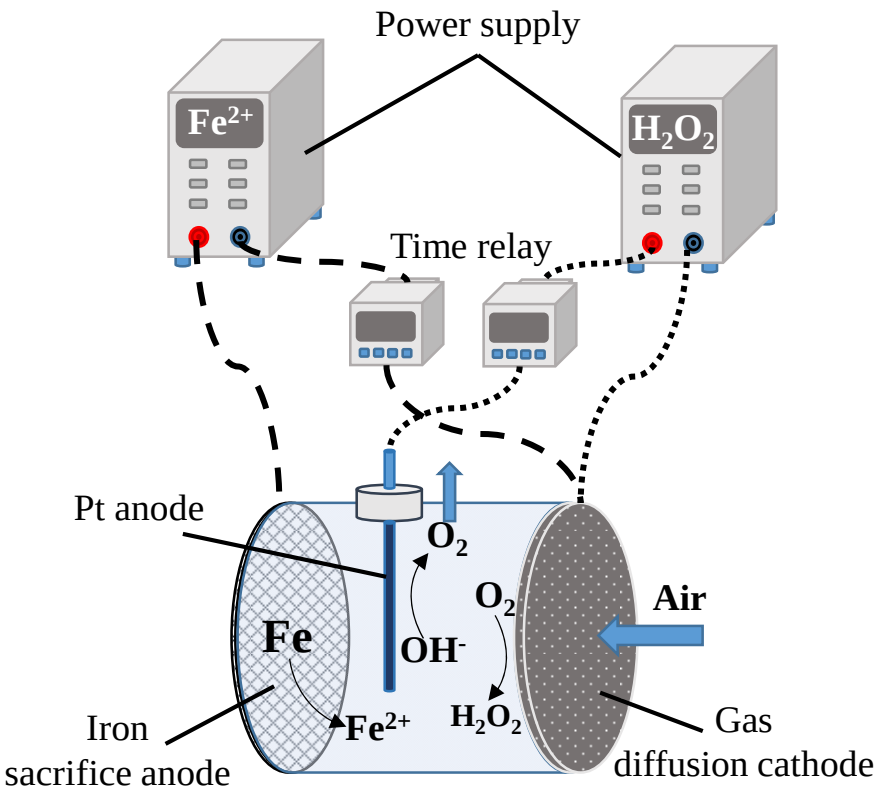

Fig. S1 Schematic diagram of the pulsed switching peroxi-coagulation system