

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19

**Enhanced removal of antibiotics in hospital wastewater by Fe-ZnO
activated persulfate-based oxidation**

Gnougouon Nina COULIBALY^{1,2}, Sungjun BAE², Joohyun KIM², Aymen Amin ASSADI¹,
Khalil HANNA^{1*}

¹Univ. Rennes, Ecole Nationale Supérieure de Chimie de Rennes, CNRS, ISCR-UMR 6226,
F-35000 Rennes, France

²Department of Civil and Environmental Engineering, Konkuk University, 120 Neungdong-ro,
Gwangjin-gu, Seoul 05029, Republic of Korea

*Corresponding author: khalil.hanna@ensc-rennes.fr

21 Tables and figures

23 **Table S1.** EDX analysis results for different samples.....3

24 **Table S2.** Inorganic species and physico-chemical characteristics of SWW and RHW.....4

26 **Fig. S1.** Schematic diagram of recirculation glass reactor system.....5

27 **Fig. S2.** EDX analysis of (a) ZnO and (b) Fe-ZnO particles.....6

28 **Fig. S3.** FLU removal kinetics in presence of oxidants.....7

29 **Fig. S4.** Removal kinetics of FLU with 0.7 wt% Fe-ZnO/CA membrane catalyst at different

30 concentration.....8

31 **Fig. S5.** Removal of (a): FLU and (b): CIP in single system in different water matrices.....9

32 **Fig. S6.** Effect of water matrices in (a): FLU and (b): CIP degradation in binary system.....10

33 **Fig. S7.** Adsorption of inorganic ligands and LHA on TiO₂ and 0.7 wt % Fe-ZnO/CA

34 membrane surface in dark, without FLU and PS.....11

Table S1. EDX analysis results for different samples

Chemical elements	Nanowire ZnO	Nanoparticles
	(wt %)	Fe-doped ZnO
Zn	80	79.04
O	20	18.05
Fe	0	2.91

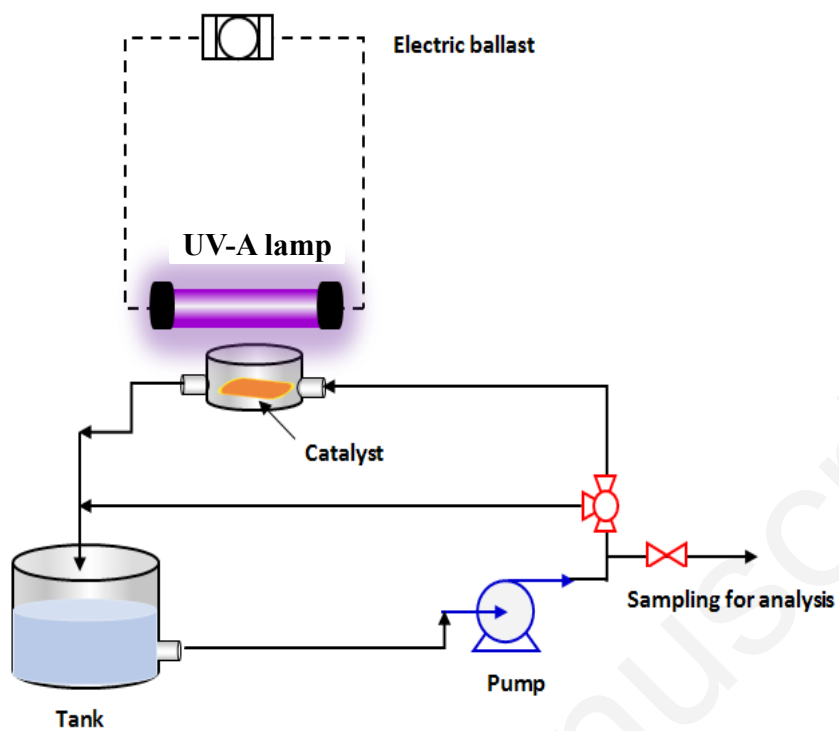
Table S2. Inorganic species and physico-chemical characteristics of SWW and RHW

	SWW	RHW
pH	8.0±0.5	6.8±0.2
Turbidity (NTU)	2±1	196±5
Conductivity ($\mu\text{S cm}^{-1}$)	1250±5	1340±5
TOC (mg L^{-1})	80±5	50±10
Suspended solid (mg L^{-1})	0	20±2
Chloride (mg L^{-1})	450±20	620±10
Nitrate (mg L^{-1})	35±2	7±2
Sulfate (mg L^{-1})	20±2	60±10
Phosphate (mg L^{-1})	150±10	60±10

Synthetic wastewater (SWW) were prepared by adding 400 mg L^{-1} of NaCl, 50 mg L^{-1} of citric acid, 30 mg L^{-1} of ascorbic acid, 100 mg L^{-1} of sucrose and 230 mg L^{-1} Na_2HPO_4 to tap water (conductivity 408 $\mu\text{S cm}^{-1}$).

17

18



73

74 **Fig. S1.** Schematic diagram of recirculation glass reactor system

75

76

77

78

79

80

81

82

83

84

85

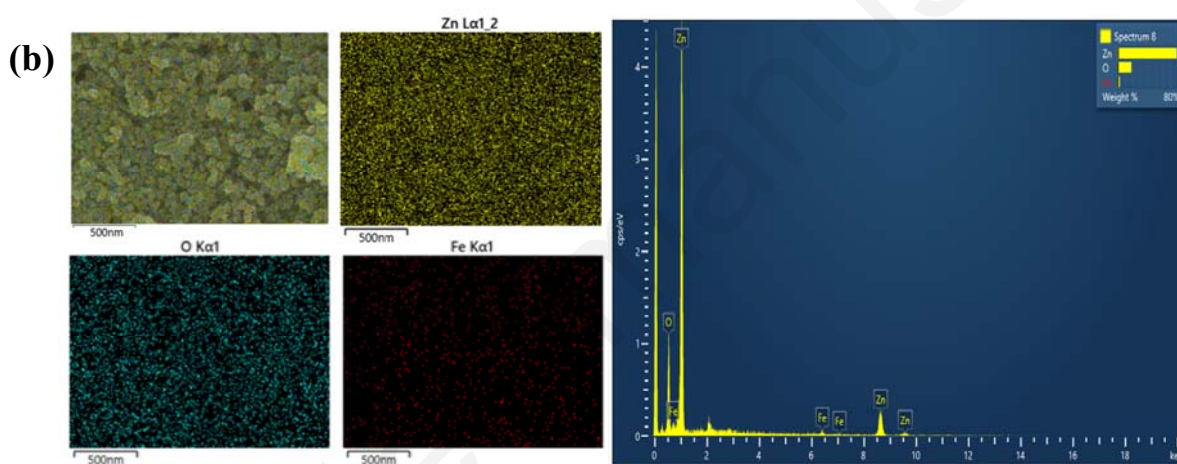
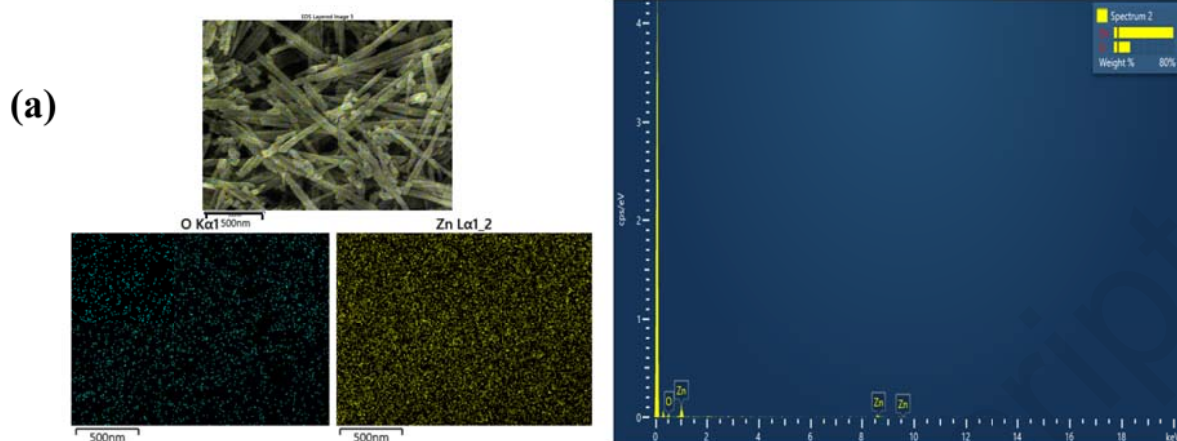
86

87

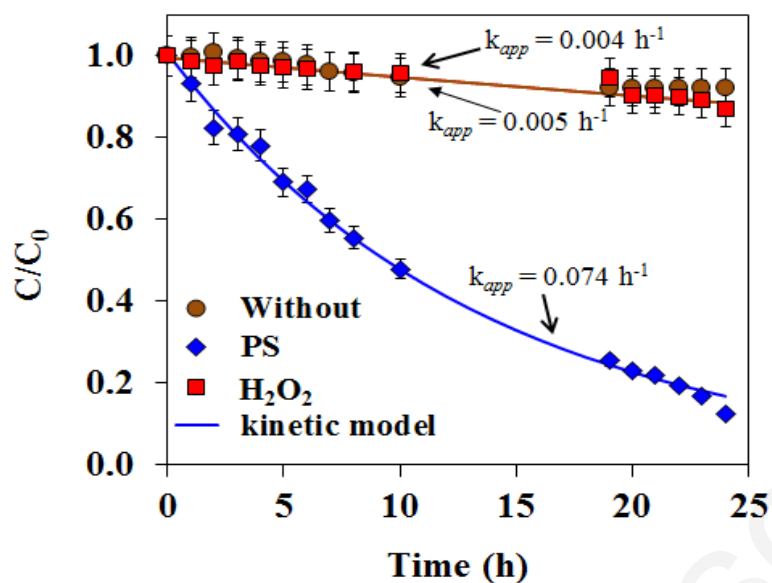
88

19

20



93**Fig. S2.** EDX analysis of (a) ZnO and (b) Fe-ZnO particles. Abbreviations: energy dispersive
94X-ray (EDX)



103

104

105 **Fig. S3.** FLU removal kinetics in presence of oxidants. Experimental conditions: $[\text{FLU}]_0 = 5$
 106 μM , $[\text{PS}]_0 = 0.5 \text{ mM}$, $[\text{H}_2\text{O}_2]_0 = 0.2 \text{ mM}$, UV-A reaction time = 24 h. $\text{pH}_0 = 7.0 \pm 0.2$, $V = 1$
 107 L, recirculation flow rate = 222 mL min^{-1} . The correlation coefficients for kinetic models were
 108 more than 0.98). Abbreviations: FLU = flumequine, PS = Persulfate, H_2O_2 = hydrogen
 109 peroxide.

110

111

112

113

114

115

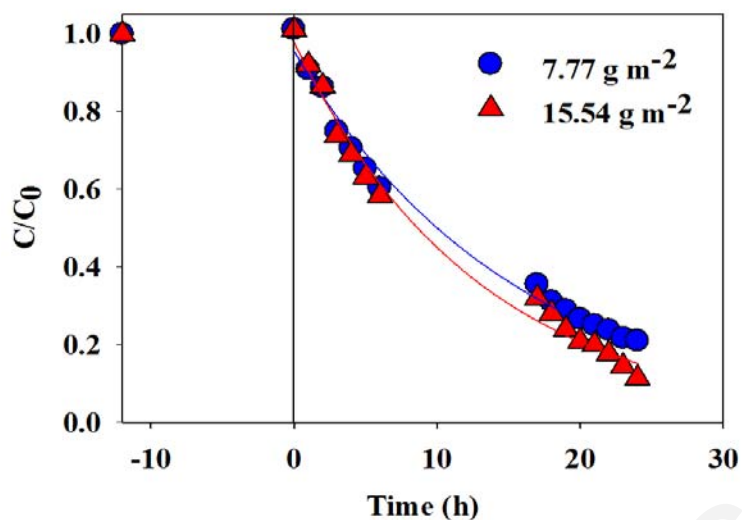
116

117

118

119

120



121

122**Fig. S4.** Removal kinetics of FLU with 0.7 wt% Fe-ZnO/CA membrane catalyst at different
123concentration. Experimental conditions: [FLU]₀ = 5 μM, UV-A reaction time = 24 h UV-A
124reaction time = 24 h. pH₀ = 7.0 ± 0.2, recirculation flow rate = 222 mL min⁻¹. The correlation
125coefficients for kinetic models were more than 0.99.

126

127

128

129

130

131

132

133

134

135

136

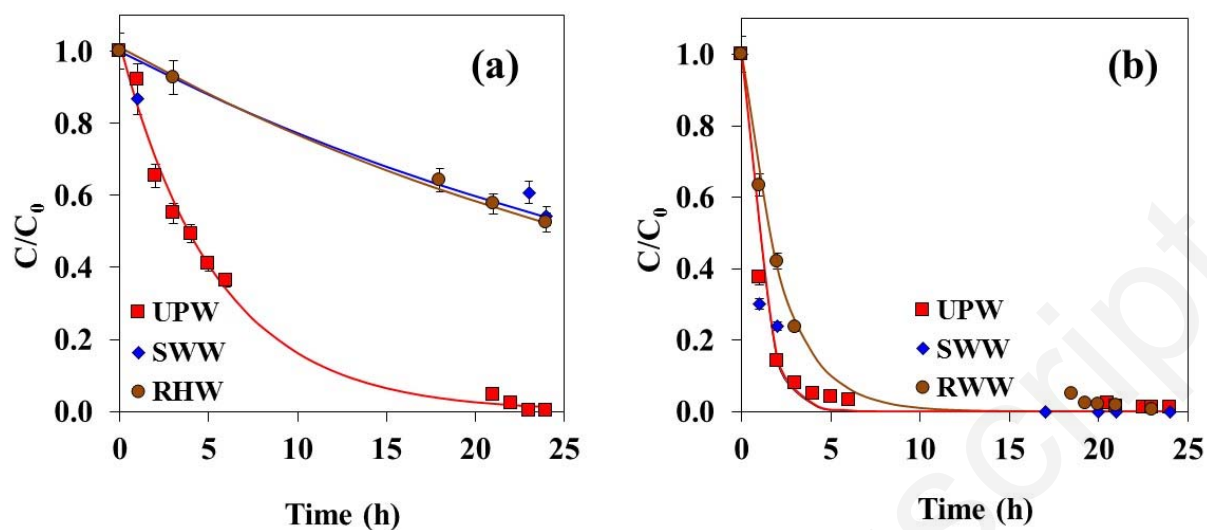
137

138

139

31

32



141

142

143 **Fig. S5.** Removal of (a): FLU and (b): CIP in single system in different water matrices.

144 Experimental conditions: $[\text{FLU}]_0 = [\text{CIP}]_0 = 5 \mu\text{M}$, $[\text{PS}]_0 = 0.5 \text{ mM}$, $[0.7 \text{ wt } \% \text{ Fe-ZnO}] =$

145 7.77 g m^{-2} CA membrane, UV-A reaction time = 24 h, $\text{pH}_0 = 7.0 \pm 0.2$, $V = 1 \text{ L}$, recirculation

146 flow rate = 222 mL min^{-1} . The correlation coefficients for kinetic models were more than

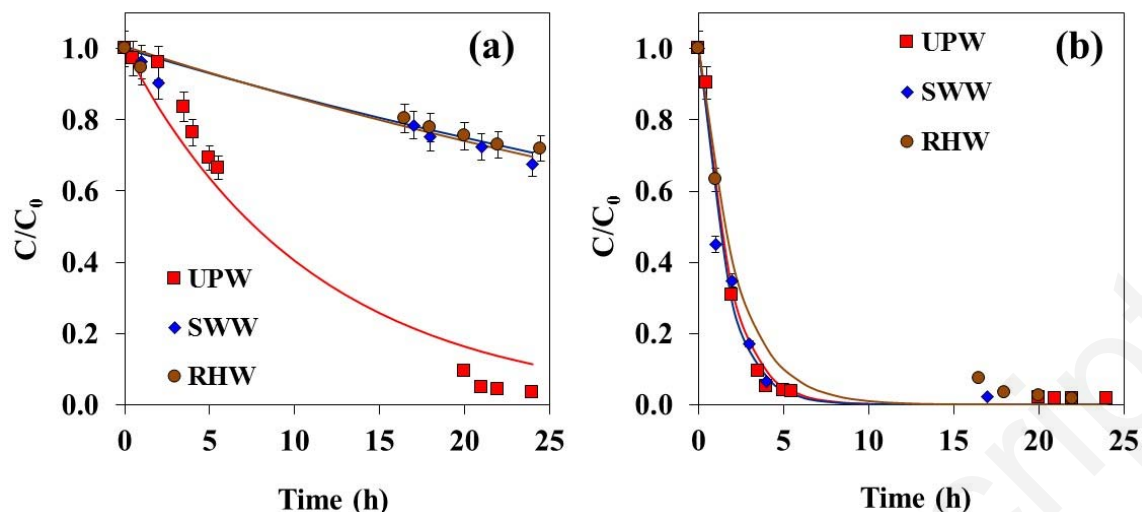
147 0.99. Abbreviations: FLU = flumequine, CIP = ciprofloxacin, PS = Persulfate: UPW = Ultra-

148 pure water, SWW = Synthetic wastewater, RHW = Real hospital wastewater.

149

150

151



152

153**Fig. S6.** Effect of water matrices in (a): FLU and (b): CIP degradation in binary system.

154Experimental conditions: $[FLU] = [CIP]_0 = 5 \mu\text{M}$, $[PS]_0 = 0.5 \text{ mM}$, $[0.7 \text{ wt } \% \text{ Fe-ZnO}] =$

1557.77 g m^{-2} CA membrane, UV-A reaction time = 24 h, $\text{pH}_0 = 7.0 \pm 0.2$, $V = 1 \text{ L}$, recirculation

156flow rate = 222 mL min^{-1} . The correlation coefficients for kinetic models were more than

1570.95. Abbreviations: FLU = flumequinone, CIP = ciprofloxacin, PS = persulfate, UPW = Ultra-

158pure water, SWW = Synthetic wastewater, RHW = Real hospital wastewater.

159

160

161

162

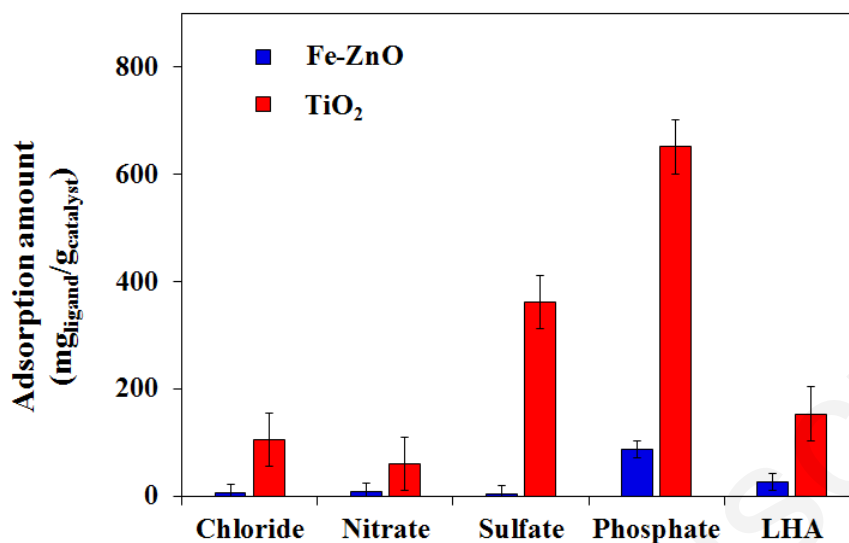
163

164

165

166

167



169

170**Fig. S7.** Adsorption of inorganic ligands and LHA on TiO₂ and 0.7 wt % Fe-ZnO/CA
171membrane surface in dark, without FLU and PS. Experimental conditions: catalyst mass =
1720.025 g, [Phosphate]₀ = 150 mg L⁻¹, [Nitrate]₀ = 10 mg L⁻¹, [Sulfate]₀ = 120 mg L⁻¹,
173[Chloride]₀ = 250 mg L⁻¹ and [LHA]₀ = 40 mgC L⁻¹, reaction time = 12 h, pH₀ = 7.0 ± 0.2, V
174= 1 L, recirculation flow rate = 222 mL min⁻¹. Abbreviation: FLU = flumequine, PS =
175Persulfate, LHA = Leonardite Humic Acid.

176

177

178

179