Sequencing batch photocatalytic H_2O_2 production over magnetic resorcinol-formaldehyde polymer for on-site water purification by UV light irradiation

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Table S1 Comparison of photocatalytic H_2O_2 production over different photocatalysts in pure water

Cat. (g/L)	Λ (nm)	Time (h)	H_2O_2 (μmol/L)	Ref.
Au/WO_3 (1.67)	>420	10	13.9	1
$Au/BiVO_4$(1.67)	>420	10	40.2	1
Co-GCN/AQ (0.5)	>300	1	60	2
Znnppc-BCN (0.4)	400-800	1	45.6	3
CdS/rGO (1.0)	>420	12	130	4
SbPCN (2.0)	>420	2	365	5
ORP/GCN (1.0)	>420	10	400	6
GCN/MTI (1.67)	420-500	24	917	7
GCN/PDI/rGO (1.67)	420-500	24	970	8
MRF (1 g/L)	**>420**	**12**	**1200**	**This work**
CoO : $MoBiVO_4$: Pd (0.17)	AM 1.5	1	1425	9
RF523 (1.67)	420-700	24	3300	10

Table S2 Removal efficiency and EEO value of different treatment processes

Treatment process	Contaminants	Removal efficiency (%)	EEO value (kWh/m^3/order)	Ref.
Photo-Fenton	Reactive Orange 4	100%	357.1	11
UV-H_2O_2	Reactive Orange 4	80%	1666.7	11
UV-TiO_2	Reactive Black 5	70%	220.0	12
UV-TiO_2	Sodium dodecylbenzenesulfonate	100%	595.0	13
UV/Peroxydisulfate	Basic Red 46	80%	155.4	14
Flow-through UV-H_2O_2	Rhodamine B	99%	10.4	This Work
	Humic acid	65%	54.8	
	Tetracycline	80%	35.8	

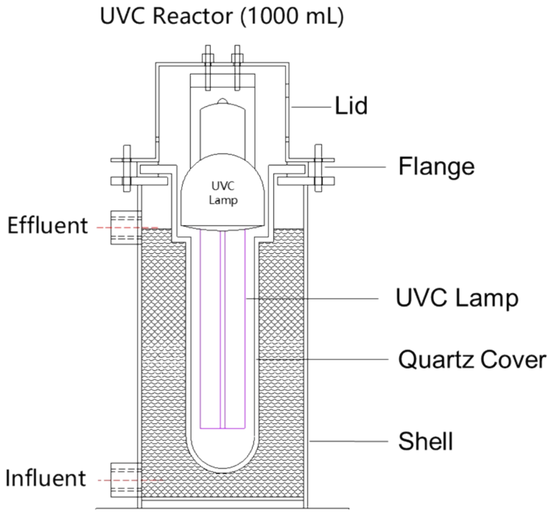

Figure S1 The section view of UVC reactor for a flow through process.

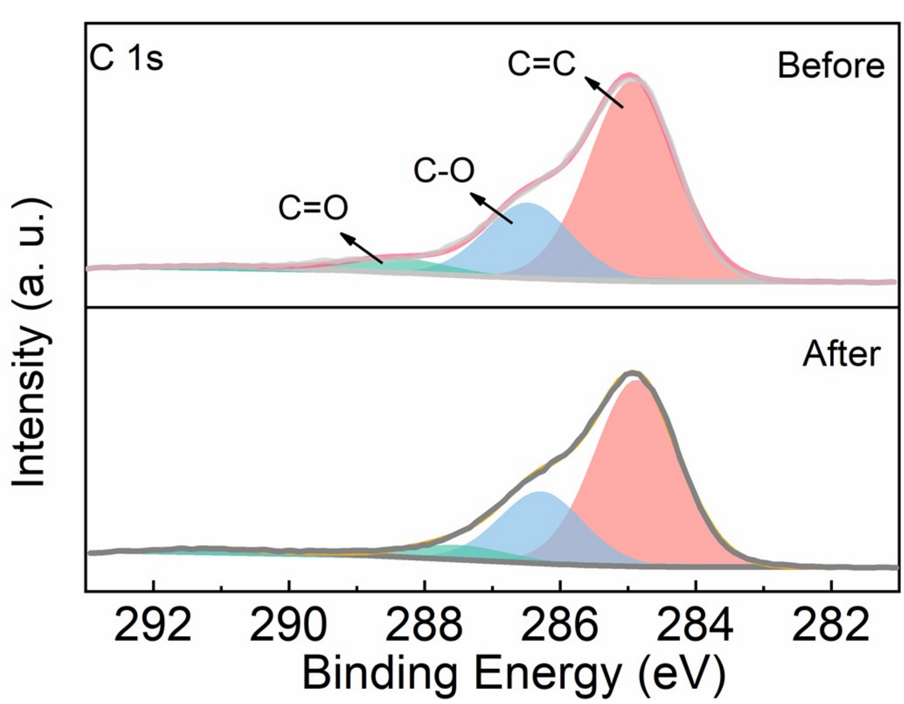

Figure S2 The C 1s high-resolution XPS spectra of MRF before and after usage.

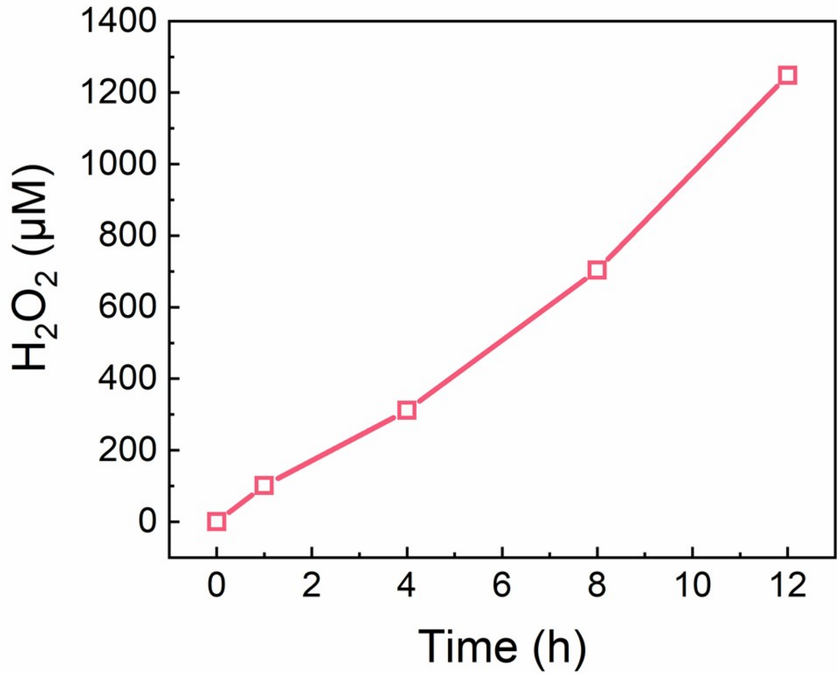

Figure S3 Photocatalytic H_2O_2 production over MRF upon prolonged irradiation (catalysts: 1 g/L, pH = 7, λ > 420 nm, Air: 0.5 L/min).

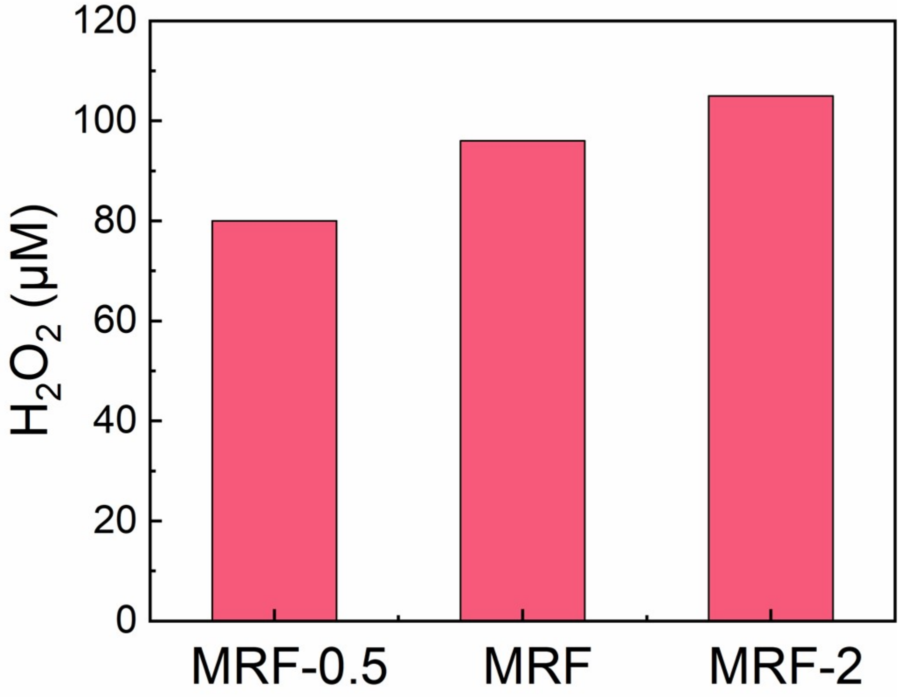

Figure S4 Photocatalytic H_2O_2 production within 60 min over MRFs with different RF contents (catalysts: 1 g/L, pH = 7.5, λ > 420 nm, Air: 0.5 L/min).

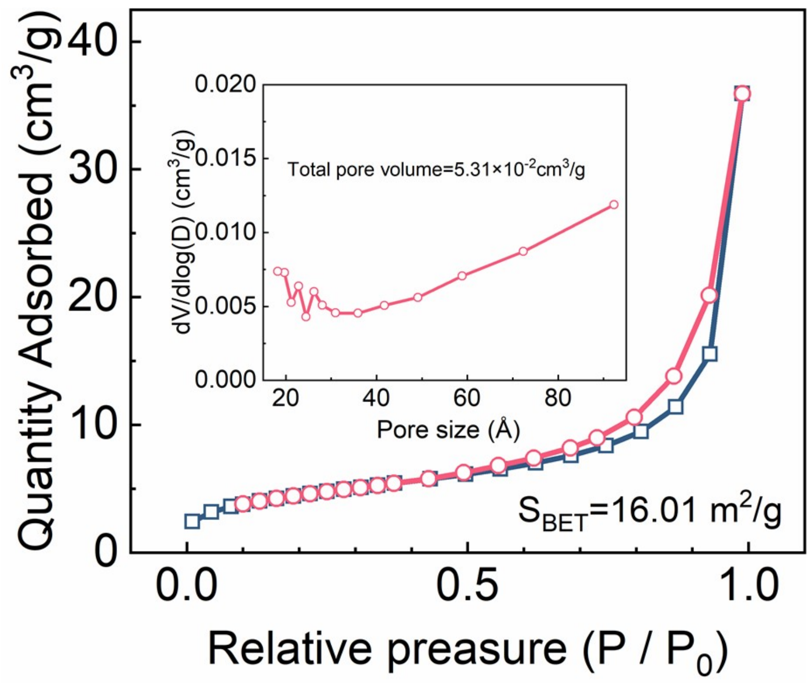

Figure S5 N_2 adsorption/desorption analysis for MRF (inset is the pore-size distribution curve).

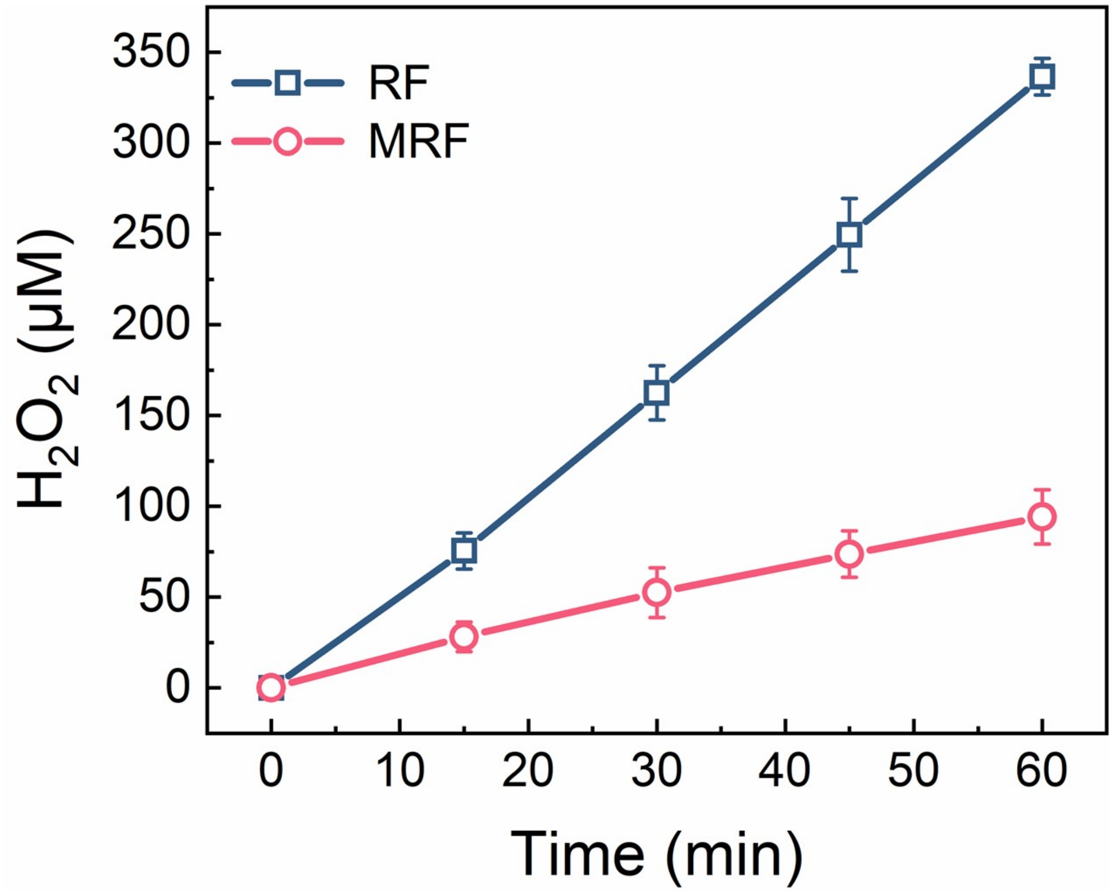

Figure S6 Photocatalytic H_2O_2 production over RF and MRF powders (Catalysts: 1 g/L, pH=7, λ>420 nm, Air: 0.5 L/min).

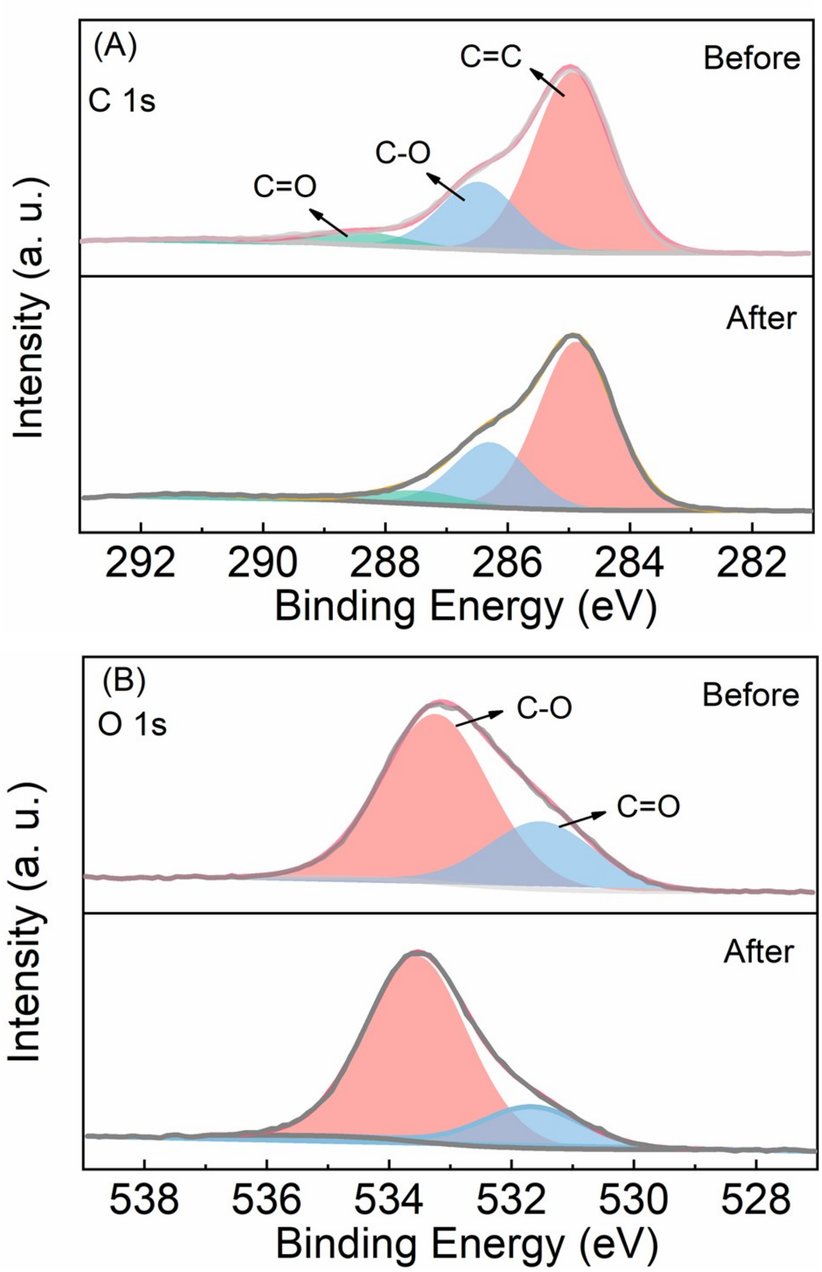

Figure S7 (A) C 1s and (B) O 1s high-resolution XPS spectra of MRF before and after photocatalytic reaction.

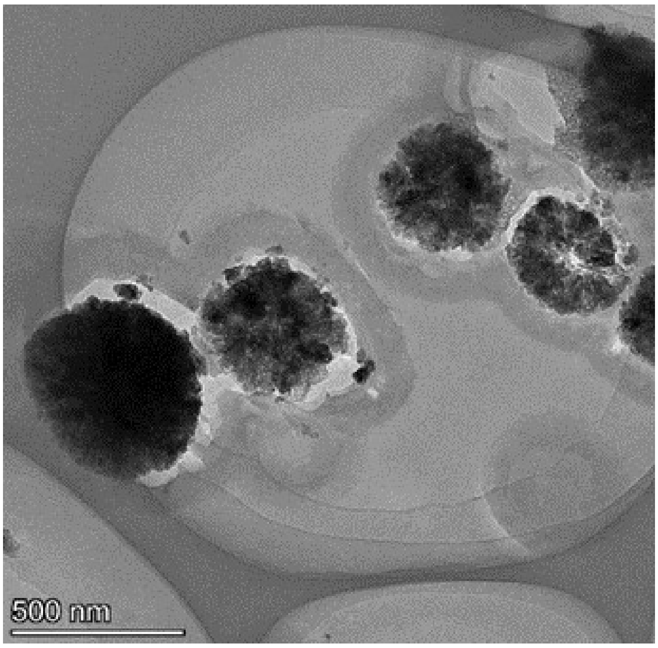

Figure S8 TEM image of MRF after photocatalytic reaction.

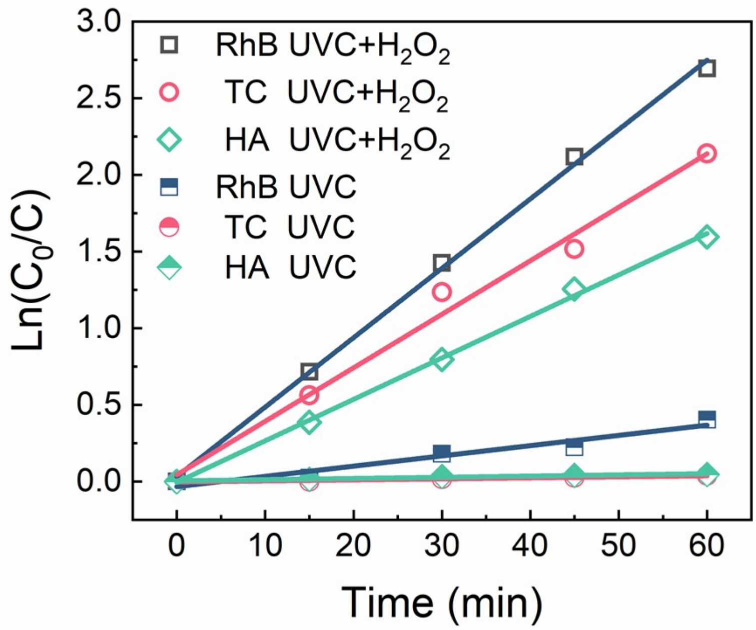

Figure S9 Kinetic model of RhB, TC and HA decomposition in the batch UVC/H_2O_2 process (RhB, TC or HA: 1 mg/L; H_2O_2: 10 μM).

The Slope and R^2 of kinetic model in **Figure S9**

	Slope (min^{-1})	R^2
RhB UVC+H_2O_2	0.045	0.997
TC UVC+H_2O_2	0.032	0.984
HA UVC+H_2O_2	0.027	0.998
RhB UVC	0.0067	0.923
TC UVC	0.00066	0.930
HA UVC	0.00078	0.932

References

1. H. Hirakawa, S. Shiota, Y. Shiraishi, H. Sakamoto, S. Ichikawa and T. Hirai, Au Nanoparticles Supported on $BiVO_4$: Effective Inorganic Photocatalysts for H_2O_2 Production from Water and O_2 under Visible Light, *ACS Catalysis*, 2016, **6**, 4976-4982.
2. C. Chu, Q. Zhu, Z. Pan, S. Gupta, D. Huang, Y. Du, S. Weon, Y. Wu, C. Muhich, E. Stavitski, K. Domen and J. H. Kim, Spatially separating redox centers on 2D carbon nitride with cobalt single atom for photocatalytic H_2O_2 production, *Proc Natl Acad Sci U S A*, 2020, **117**, 6376-6382.
3. Y. X. Ye, J. Pan, F. Xie, L. Gong, S. Huang, Z. Ke, F. Zhu, J. Xu and G. Ouyang, Highly efficient photosynthesis of hydrogen peroxide in ambient conditions, *Proc Natl Acad Sci U S A*, 2021, **118**.
4. S. Thakur, T. Kshetri, N. H. Kim and J. H. Lee, Sunlight-driven sustainable production of hydrogen peroxide using a CdS-graphene hybrid photocatalyst, *J. Catal.*, 2017, **345**, 78-86.
5. Z. Teng, Q. Zhang, H. Yang, K. Kato, W. Yang, Y.R. Lu, S. Liu, C. Wang, A. Yamakata, C. Su, B. Liu and T. Ohno, Atomically dispersed antimony on carbon nitride for the artificial photosynthesis of hydrogen peroxide, *Nature Catalysis*, 2021, **4**, 374-384.
6. J. Zhang, J. Lang, Y. Wei, Q. Zheng, L. Liu, Y.H. Hu, B. Zhou, C. Yuan and M. Long, Efficient photocatalytic H_2O_2 production from oxygen and pure water over graphitic carbon nitride decorated by oxidative red phosphorus, *Applied Catalysis B: Environmental*, 2021, **298**.
7. Y. Kofuji, S. Ohkita, Y. Shiraishi, H. Sakamoto, S. Ichikawa, S. Tanaka and T. Hirai, Mellitic Triimide-Doped Carbon Nitride as Sunlight-Driven Photocatalysts for Hydrogen Peroxide Production, *ACS Sustainable Chemistry & Engineering*, 2017, **5**, 6478-6485.
8. Y. Kofuji, Y. Isobe, Y. Shiraishi, H. Sakamoto, S. Tanaka, S. Ichikawa and T. Hirai, Carbon Nitride-Aromatic Diimide-Graphene Nanohybrids: Metal-Free Photocatalysts for Solar-to-Hydrogen Peroxide Energy Conversion with 0.2% Efficiency, *Journal of the American Chemical Society*, 2016, **138**, 10019-10025.
9. T. Liu, Z. Pan, J. J. M. Vequizo, K. Kato, B. Wu, A. Yamakata, K. Katayama, B. Chen, C. Chu and K. Domen, Overall photosynthesis of H_2O_2 by an inorganic semiconductor, *Nature communications*, 2022, **13**, 1034.
10. Y. Shiraishi, T. Takii, T. Hagi, S. Mori, Y. Kofuji, Y. Kitagawa, S. Tanaka, S. Ichikawa and T. Hirai, Resorcinol-formaldehyde resins as metal-free semiconductor photocatalysts for solar-to-hydrogen peroxide energy conversion, *Nature Materials*, 2019, **18**, 985-993.
11. M. Muruganandham, K. Selvam and M. Swaminathan, A comparative study of quantum yield and electrical energy per order EEo for advanced oxidative decolourisation of reactive azo dyes by UV light, *J Hazard Mater*, 2007, **144**, 316-322.

12. L. C. Ferreira, M. S. Lucas, J. R. Fernandes and P. B. Tavares, Photocatalytic oxidation of Reactive Black 5 with UV-A LEDs, *Journal of Environmental Chemical Engineering*, 2016, **4**, 109-114.
13. S. Dominguez, M. J. Rivero, P. Gomez, R. Ibañez and I. Ortiz, Kinetic modeling and energy evaluation of sodium dodecylbenzenesulfonate photocatalytic degradation in a new LED reactor, *Journal of Industrial and Engineering Chemistry*, 2016, **37**, 237-242.
14. M. H. Rasoulifard, R. Marandi, H. Majidzadeh and I. Bagheri, Ultraviolet Light-Emitting Diodes and Peroxydisulfate for Degradation of Basic Red 46 from Contaminated Water, *Environmental Engineering Science*, 2011, **28**, 229-235.