Electronic Supplementary Material (ESI) for Environmental Science: Water Research & Technology. This journal is © The Royal Society of Chemistry 2020

Supplementary information

Dissolved organic matter modified magnetic carbon nanotubes enhance the

bioremediation of azo dyes and Cr(VI)

Caiwen He^a, Lipeng Gu^b, Huan He^{*,a,c}, Zhe Zhang^c, Xiaoxia Wang^a, Fengxia Han^{**,a} Bin

Huang^{a,d}, and Xuejun Pan^{a,d}

Fig. S1. Nitrogen adsorption-desorption isotherms curves of AQS/Fe₃O₄/CNTs and

HA/Fe₃O₄/CNTs

Fig. S2. Fluorescence spectra of AQS/Fe₃O₄/CNTs and HA/Fe₃O₄/CNTs

AQS contained model humic acid polymers, however, HA contained fulvic acid, humic acid and humic acid-like components, therefore, AQS was pure whereas HA contained complicated impurities.

Fig. S3. The cyclic voltammetry (CV) curves of Cr(VI) and methyl orange.

It was found that the reduction of Cr(VI) took precedence over methyl orange decolorization driven by anaerobic bacteria. Because as terminal electron acceptors, Cr(VI) which had relatively active redox activity received electrons in preference to methyl orange according to the cyclic voltammetry (CV) curves.

Fig. S4. The Shannon-weaver index of anaerobic bacteria (AB: bacteria in system with the addition of AQS/Fe₃O₄/CNTs, HB: bacteria in system with the addition of HA/Fe₃O₄/CNTs).

Bacterial community of HB was more diverse than AB, implying that the kind of DOM would affect the biodiversity of the anaerobic bacteria.

Table S1. The significance level of different materials.

Table S2. Effect of Fe₃O₄ on significance level of different groups.

Table S3. Effect of initial Cr(VI) concentration on significance level of different groups.

Table S4. The significance level of single system and binary system.



Fig. S1. Nitrogen adsorption-desorption isotherms curves of AQS/Fe₃O₄/CNTs and HA/Fe₃O₄/CNTs.



Fig. S2. Fluorescence spectra of AQS/Fe₃O₄/CNTs and HA/Fe₃O₄/CNTs.



Voltage /V vs Ag/AgCl Fig. S3. The cyclic voltammetry (CV) curves of Cr(VI) and methyl orange.



Fig. S4. The Shannon-weaver index of anaerobic bacteria (AB: bacteria in system with the addition of AQS/Fe₃O₄/CNTs, HB: bacteria in system with the addition of HA/Fe₃O₄/CNTs).

Table S1. The significance level of different materials.

(J) Material	t (h)	<i>p</i> Cr(VI)	p MO
	6	0.001	0.116
	12	0.023	0.774
HA/Fe ₃ O ₄ /CNTs-0.8	24	0.007	0.339
	30	0.056	0.220
	36	0.002	0
	(J) Material HA/Fe ₃ O ₄ /CNTs-0.8	(J) Material t (h) 6 12 HA/Fe ₃ O ₄ /CNTs-0.8 24 30 36	(J) Material t (h) p Cr(VI) 6 0.001 12 0.023 HA/Fe ₃ O ₄ /CNTs-0.8 24 0.007 30 0.056 36 0.002

MO, methyl orange. $p \leq 0.05$ represent significant differences.

(I) Matariala	(I) Matarials	<i>p</i> (6 h)		<i>p</i> (12 h)		<i>p</i> (24 h)		<i>p</i> (30 h)		<i>p</i> (36 h)	
(1) Waterials	(J) Materials	Cr(VI)	MO	Cr(VI)	MO	Cr(VI)	МО	Cr(VI)	МО	Cr(VI)	MO
	AQS/CNTs-0.5	0.379	0.457	0.545	0.455	0.095	0.099	0.157	0.009	0.026	0.009
AQS/Fe ₃ O ₄ /CNTs- 0.5	HA/Fe ₃ O ₄ /CNTs-0.8	0	0.617	0.001	0.924	0.007	0.282	0.028	0.092	0.079	0.039
	HA/CNTs-0.8	0	0.326	0	0.042	0.001	0.014	0	0	0	0
	Fe ₃ O ₄ /CNTs	0	0.335	0	0.029	0	0.003	0	0	0	0
AQS/CNTs-0.5	HA/Fe ₃ O ₄ /CNTs-0.8	0	0.226	0	0.511	0.153	0.511	0.323	0.204	0.522	0.404
	HA/CNTs-0.8	0	0.101	0	0.151	0.010	0.282	0.001	0.003	0.028	0
	Fe ₃ O ₄ /CNTs	0	0.104	0	0.108	0.001	0.062	0	0	0	0
HA/Fe ₃ O ₄ /CNTs-0.8	HA/CNTs-0.8	0.185	0.617	0.011	0.049	0.138	0.099	0.003	0	0.009	0
	Fe ₃ O ₄ /CNTs	0.028	0.630	0.001	0.034	0.010	0.019	0	0	0	0
HA/CNTs-0.8	Fe ₃ O ₄ /CNTs	0.282	0.985	0.153	0.837	0.151	0.359	0.024	0.154	0.003	0.015

Table S2. Effect of Fe_3O_4 on significance level of different groups.

MO, methyl orange. $p \leq 0.05$ represent significant differences.

(I)	(I) (J)		<i>p</i> (6 h)		<i>p</i> (12 h)		<i>p</i> (24 h)		<i>p</i> (30 h)		<i>p</i> (36 h)	
Initial Cr(VI) concentration	Initial Cr(VI) concentration	Cr(VI)	МО	Cr(VI)	МО	Cr(VI)	МО	Cr(VI)	МО	Cr(VI)	МО	
$(mg L^{-1})$	$(mg L^{-1})$											
10	15	0	0.231	0	0.112	0.002	0.002	0	0.001	0	0	
(AQS/Fe ₃ O ₄ /CNTs	20	0	0.359	0	0.130	0	0	0	0	0	0	
)	25	0	0.087	0	0.073	0	0	0	0	0	0	
15	20	0.663	0.754	0.312	0.928	0.035	0.002	0	0.023	0	0	
(AQS/Fe ₃ O ₄ /CNTs)	25	0.125	0.535	0.100	0.785	0.008	0.001	0	0.009	0	0	
20 (AQS/Fe ₃ O ₄ /CNTs)	25	0.242	0.359	0.458	0.717	0.381	0.561	0.085	0.564	0.101	0.232	
10	15	0.033	0.273	0.008	0.001	0.002	0.009	0	0	0	0	
$\frac{10}{(114/E_{\odot} O / ONE_{\odot})}$	20	0.027	0.455	0.003	0	0	0.002	0	0	0	0	
$(HA/Fe_3O_4/CNTS)$	25	0.015	0.086	0.001	0	0	0.002	0	0	0	0	
15	20	0.901	0.705	0.491	0.218	0.028	0.349	0.006	0.016	0	0.007	
(HA/Fe ₃ O ₄ /CNTs)	25	0.621	0.455	0.182	0.147	0.013	0.279	0.002	0.007	0	0.003	
20 (HA/Fe ₃ O ₄ /CNTs)	250	0.710	0.273	0.480	0.796	0.625	0.872	0.358	0.625	0.317	0.654	

Table S3. Effect of initial Cr(VI) concentration on significance level of different groups.

MO, methyl orange. $p \leq 0.05$ represent significant differences.

(I)	(J)		<i>p</i> (A	QS/Fe ₃ O ₄ /C	NTs)		p (HA/Fe ₃ O ₄ /CNTs)					
Systems	Systems	6 h	12 h	24 h	30 h	36 h	6 h	12 h	24 h	30 h	36 h	
Cr(VI)-S	Cr(VI)-B	0.063	0.394	0.006	0.011	1.000	0.042	0.400	0.046	0	0.002	
MO-S	MO-B	0.002	0.012	0.002	0.008	0.286	0.003	0.044	0.003	0	0.002	

Table S4. The significance level of single system and binary system.

S, single pollutant system with Cr(VI) or methyl orange alone; B, binary pollutants system with Cr(VI) or methyl orange in combination. MO, methyl orange. $p \leq 0.05$ represent significant differences.