Supporting Information

Double-network hydrogel templated Fe/graphene with enhanced PMS activation performance: considering the effect of template and iron species

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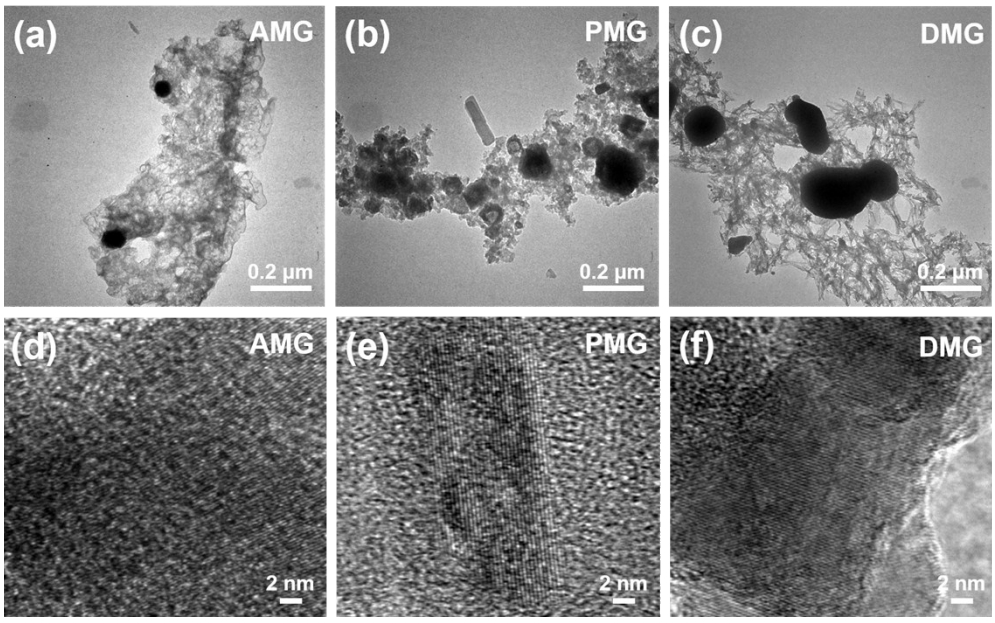


Figure S1 HRTEM of AMG (a and d), PMG (b and e) and DMG (c and f).

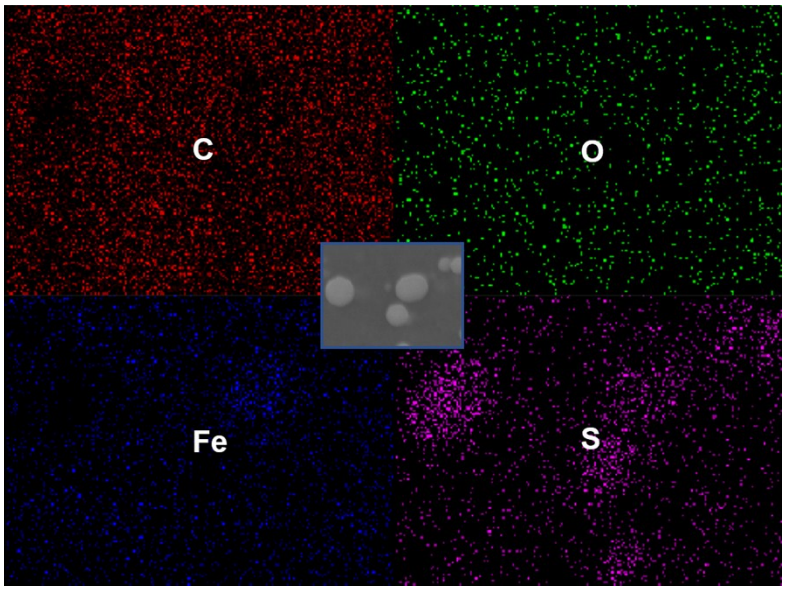
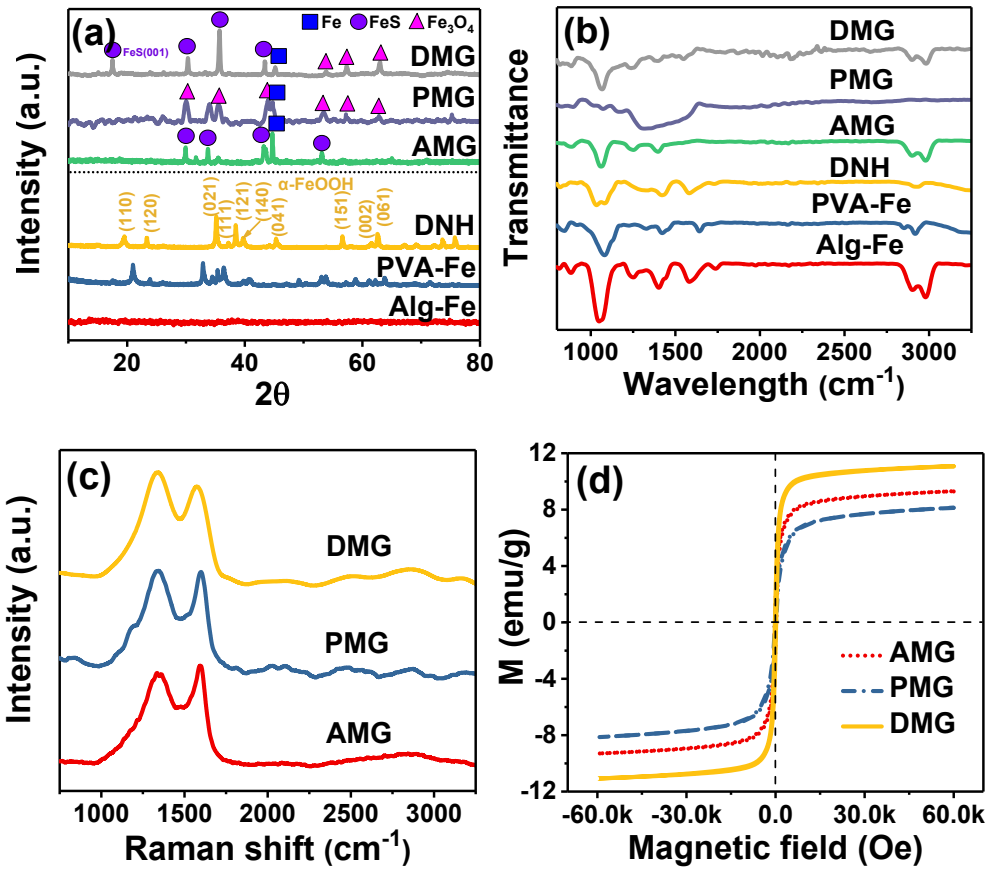


Figure S2 SEM-EDS mapping of DMG.



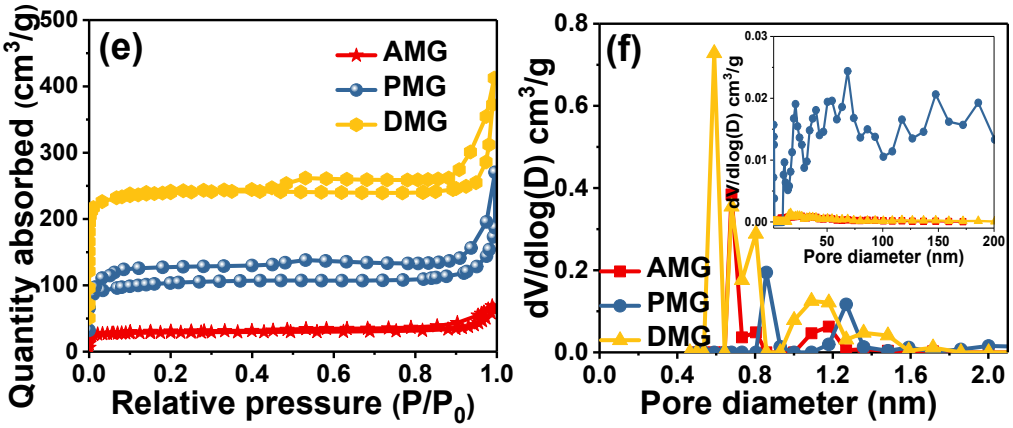


Figure S3 (a) XRD patterns of Alg-Fe, PVA-Fe, DNH, AMG, PMG and DMG, (b) FTIR spectra of Alg-Fe, PVA-Fe, DNH, AMG, PMG and DMG, (c) Raman spectra of AMG, PMG and DMG, (d) magnetization curves of AMG, PMG and DMG, (e) N_2 adsorption-desorption isotherms of AMG, PMG and DMG, (f) pore distributions of AMG, PMG and DMG.

Table S1 BET results for AMG, PMG and DMG.

Material	Surface area (m^2/g)	Pore volume (cm^3/g)	Pore size (nm)
AMG	112.21	0.02	4.21
PMG	145.94	0.05	4.43
DMG	218.75	0.06	5.24

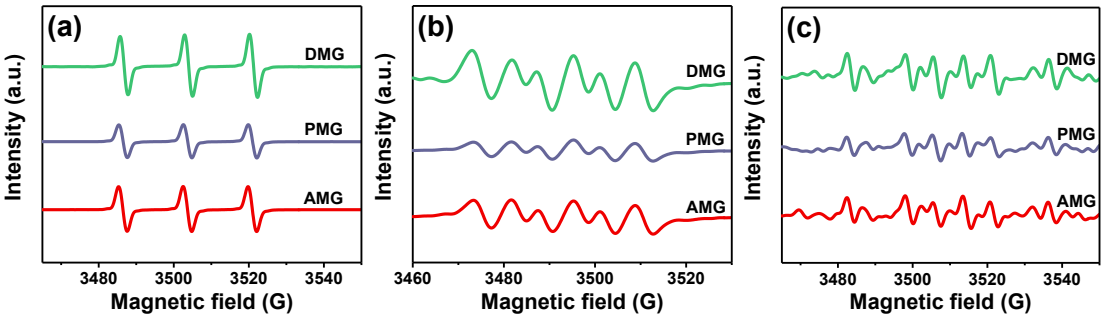
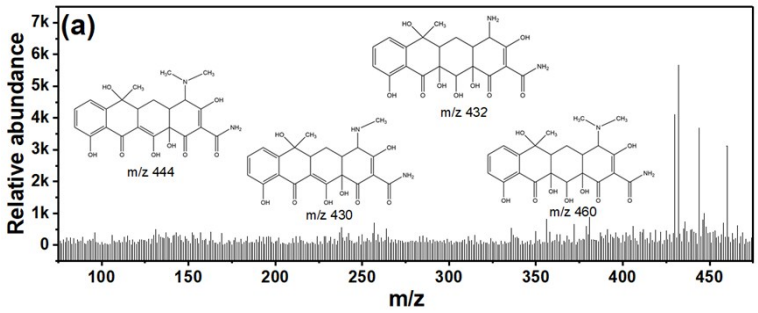
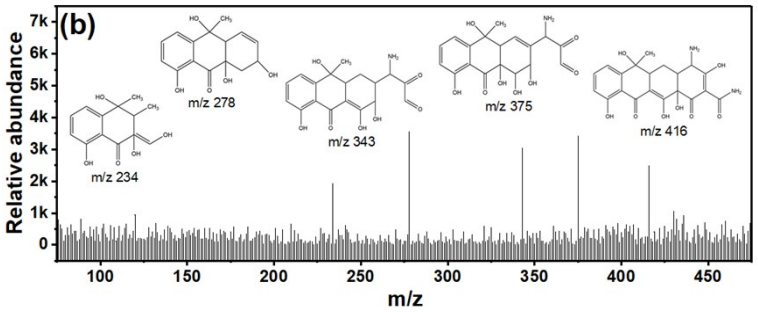
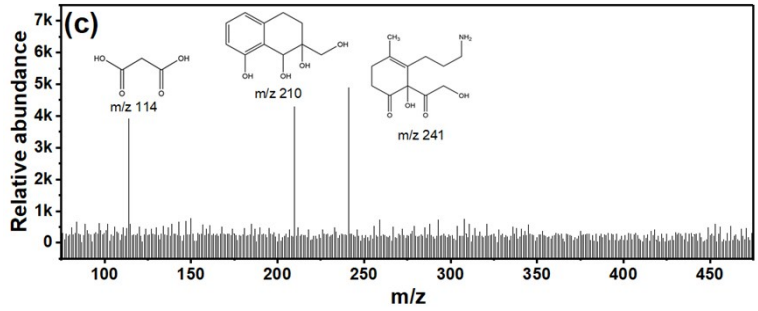


Figure S4 ESR spectra. (a) TEMP-$^1O^2$, (b) DMPO-$O_2^{\bullet-}$, (c) DMPO-$^{\bullet}$OH and DMPO-$SO_4^{\bullet-}$.







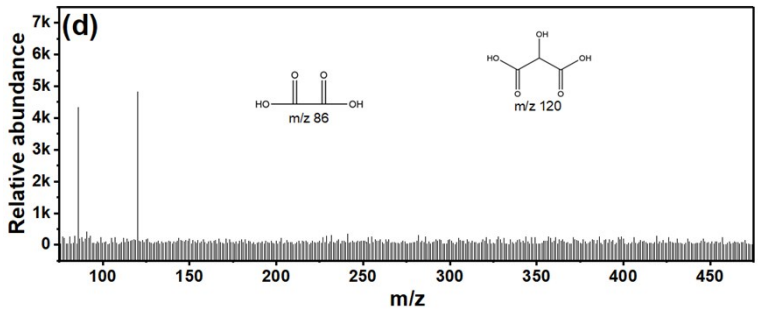


Figure S5 Mass spectra of compounds derived from UPLC-Q-TOF MS analysis of tetracycline degradation in DMG/PMS system.

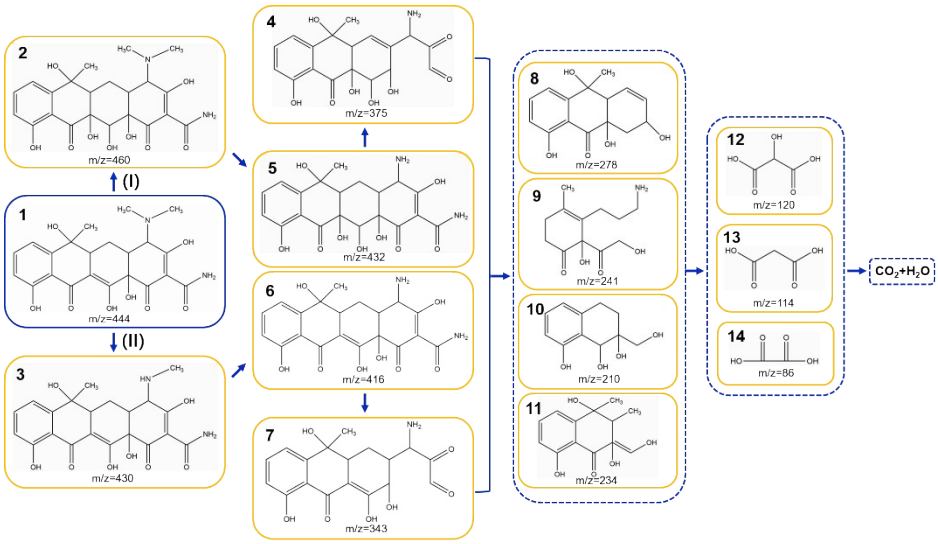


Figure S6 Two possible transformation pathways of tetracycline degradation in the DMG/PMS system.

Table S2 Condensed local electrophilicity/nucleophilicity index of tetracycline.

Atom	Electrophilicity	Nucleophilicity
1(C)	0.16035	0.00182
2(C)	0.08286	0.00157
3(C)	0.10911	0.00145

4(C)	0.06533	0.00054
5(C)	0.0957	0.00017
6(C)	0.06615	0.00143
7(C)	0.31602	0.00347
8(C)	0.06642	0.00630
9(C)	0.01353	0.00479
10(C)	0.01383	0.00176
11(C)	0.19324	-0.00085
12(C)	0.02359	0.00035
13(C)	0.00808	0.00747
14(C)	0.00987	0.00992
15(C)	0.00638	0.00820
16(C)	0.00687	0.03524
17(C)	0.00775	0.01874
18(C)	0.0037	0.03900
19(O)	0.0879	0.00170
20(O)	0.03286	0.00265
21(C)	0.01303	0.00123
22(O)	0.32425	0.00615
23(O)	0.12554	0.00686
24(N)	0.00184	0.39614
25(C)	0.0032	0.08293
26(C)	0.00706	0.07563
27(O)	0.00963	0.02520
28(C)	0.00395	0.01269

29(O)	0.00689	0.01861
30(N)	0.00338	0.01061
31(O)	0.01773	0.03224
32(O)	0.04028	0.01366
33(H)	0.06196	0.00095
34(H)	0.04186	0.00093
35(H)	0.03448	0.00090
36(H)	0.02696	0.00697
37(H)	0.0189	0.02221
38(H)	0.01421	0.01537
39(H)	0.01848	0.01126
40(H)	0.00858	0.04768
41(H)	0.04532	0.00089
42(H)	0.01977	0.00207
43(H)	0.01484	0.00146
44(H)	0.01159	0.00119
45(H)	0.0133	0.00193
46(H)	0.03672	0.00440
47(H)	0.0029	0.06526
48(H)	0.00284	0.05375
49(H)	0.0032	0.1101
50(H)	0.0077	0.05899
51(H)	0.0053	0.04504
52(H)	0.00522	0.10464
53(H)	0.00415	0.01574

54(H)	0.00267	0.00772
55(H)	0.00255	0.00543
56(H)	0.03101	0.00755

Table S3 Hirshfeld charges, condensed Fukui functions and condensed dual descriptors of tetracycline.

Atom	q(N)	q(N+1)	q(N-1)	f-	f+	f0	CDD
1(C)	-0.0296	-0.0976	-0.0284	0.0013	0.0679	0.0346	0.0666
2(C)	-0.0678	-0.1029	-0.0667	0.0011	0.0351	0.0181	0.0340
3(C)	0.0794	0.0332	0.0804	0.0010	0.0462	0.0236	0.0452
4(C)	-0.0533	-0.0810	-0.0529	0.0004	0.0277	0.014	0.0273
5(C)	-0.0042	-0.0447	-0.0041	0.0001	0.0405	0.0203	0.0404
6(C)	-0.0611	-0.0892	-0.0601	0.0010	0.0280	0.0145	0.0270
7(C)	0.1341	0.0003	0.1366	0.0024	0.1339	0.0682	0.1314
8(C)	-0.0653	-0.0934	-0.0608	0.0044	0.0281	0.0163	0.0237
9(C)	-0.0262	-0.0320	-0.0229	0.0034	0.0057	0.0046	0.0024
10(C)	0.0837	0.0778	0.0849	0.0012	0.0059	0.0036	0.0046
11(C)	0.0980	0.0161	0.0974	-0.0006	0.0819	0.0406	0.0825
12(C)	0.0779	0.0679	0.0781	0.0002	0.0100	0.0051	0.0097
13(C)	-0.0134	-0.0168	-0.0081	0.0053	0.0034	0.0043	-0.0018
14(C)	-0.0482	-0.0524	-0.0413	0.0070	0.0042	0.0056	-0.0028
15(C)	0.1549	0.1522	0.1607	0.0058	0.0027	0.0042	-0.0031
16(C)	-0.0687	-0.0716	-0.0439	0.0248	0.0029	0.0139	-0.0219
17(C)	0.1403	0.1370	0.1535	0.0132	0.0033	0.0082	-0.0099
18(C)	0.0267	0.0251	0.0541	0.0275	0.0016	0.0145	-0.0259

19(O)	-0.2342	-0.2714	-0.233	0.0012	0.0372	0.0192	0.0360
20(O)	-0.2633	-0.2773	-0.2615	0.0019	0.0139	0.0079	0.0121
21(C)	-0.0881	-0.0936	-0.0872	0.0009	0.0055	0.0032	0.0047
22(O)	-0.2919	-0.4293	-0.2876	0.0043	0.1373	0.0708	0.1330
23(O)	-0.2060	-0.2592	-0.2012	0.0048	0.0532	0.0290	0.0483
24(N)	-0.0979	-0.0987	0.1810	0.2790	0.0008	0.1399	-0.2782
25(C)	-0.0420	-0.0433	0.0164	0.0584	0.0014	0.0299	-0.057
26(C)	-0.0487	-0.0517	0.0045	0.0533	0.0030	0.0281	-0.0503
27(O)	-0.1466	-0.1506	-0.1288	0.0178	0.0041	0.0109	-0.0137
28(C)	0.1521	0.1504	0.1610	0.0089	0.0017	0.0053	-0.0073
29(O)	-0.4116	-0.4145	-0.3985	0.0131	0.0029	0.0080	-0.0102
30(N)	-0.1385	-0.1399	-0.1310	0.0075	0.0014	0.0045	-0.0060
31(O)	-0.2185	-0.2260	-0.1958	0.0227	0.0075	0.0151	-0.0152
32(O)	-0.2367	-0.2538	-0.2271	0.0096	0.0171	0.0133	0.0074
33(H)	0.0648	0.0386	0.0655	0.0007	0.0262	0.0135	0.0256
34(H)	0.0598	0.0421	0.0605	0.0007	0.0177	0.0092	0.0171
35(H)	0.0554	0.0408	0.0560	0.0006	0.0146	0.0076	0.0140
36(H)	0.0303	0.0189	0.0352	0.0049	0.0114	0.0082	0.0065
37(H)	0.0571	0.0491	0.0728	0.0156	0.0080	0.0118	-0.0076
38(H)	0.0246	0.0186	0.0354	0.0108	0.0060	0.0084	-0.0048
39(H)	0.0465	0.0387	0.0544	0.0079	0.0078	0.0079	-0.0001
40(H)	0.0658	0.0622	0.0994	0.0336	0.0036	0.0186	-0.0299
41(H)	0.1414	0.1222	0.1420	0.0006	0.0192	0.0099	0.0186
42(H)	0.1822	0.1739	0.1837	0.0015	0.0084	0.0049	0.0069
43(H)	0.0413	0.0351	0.0424	0.00100	0.0063	0.0037	0.0053
44(H)	0.0446	0.0397	0.0454	0.0008	0.0049	0.0029	0.0041
45(H)	0.0442	0.0386	0.0456	0.0014	0.0056	0.0035	0.0043
46(H)	0.1432	0.1277	0.1463	0.0031	0.0156	0.0093	0.0125

47(H)	0.0463	0.0451	0.0923	0.0460	0.0012	0.0236	-0.0447
48(H)	0.0495	0.0483	0.0873	0.0379	0.0012	0.0195	-0.0366
49(H)	0.0267	0.0254	0.1043	0.0775	0.0014	0.0394	-0.0762
50(H)	0.0494	0.0461	0.0909	0.0415	0.0033	0.0224	-0.0383
51(H)	0.0258	0.0236	0.0576	0.0317	0.0022	0.0170	-0.0295
52(H)	0.0228	0.0206	0.0965	0.0737	0.0022	0.0380	-0.0715
53(H)	0.2212	0.2195	0.2323	0.0111	0.0018	0.0064	-0.0093
54(H)	0.1556	0.1545	0.1610	0.0054	0.0011	0.0033	-0.0043
55(H)	0.1217	0.1206	0.1255	0.0038	0.0011	0.0024	-0.0027
56(H)	0.1944	0.1812	0.1997	0.0053	0.0131	0.0092	0.0078

Table S4 Condensed local electrophilicity/nucleophilicity index of tetracycline intermediate (molecular-2).

Atom	Electrophilicity	Nucleophilicity
1(C)	0.71632	0.00001
2(C)	-0.26820	-0.00001
3(C)	1.16758	0.00001
4(C)	-0.71767	-0.00005
5(C)	0.35288	-0.00002
6(C)	-0.19657	-0.00002
7(C)	3.14726	-0.00001
8(C)	0.01268	-0.00002
9(C)	0.04951	-0.00013
10(C)	-0.00321	-0.00007
11(C)	0.03490	-0.00014
12(C)	0.01767	-0.00036

13(C)	0.00752	-0.00059
14(C)	0.00946	0.00012
15(C)	-0.00499	-0.00030
16(C)	0.00588	-0.00223
17(C)	0.00172	0.00046
18(C)	0.00307	0.00154
19(O)	0.50766	-0.00005
20(O)	0.16099	-0.00022
21(C)	0.00656	-0.00001
22(O)	2.58778	-0.00021
23(O)	0.16461	-0.00037
24(N)	0.00756	-0.03316
25(C)	0.00154	-0.00036
26(C)	0.00149	0.00001
27(O)	0.01275	-0.00257
28(C)	0.00006	-0.00004
29(O)	0.01059	-0.00104
30(N)	0.00126	-0.00033
31(O)	0.06975	-0.00208
32(O)	0.05448	-0.0008
33(O)	0.44796	-0.00021
34(H)	0.09192	0.00000
35(H)	0.01869	0.00000
36(H)	0.01304	0.00000
37(H)	0.03049	-0.00011
38(H)	0.05735	-0.00012
39(H)	0.01210	-0.00055
40(H)	0.01312	-0.00041

41(H)	0.01280	-0.00023
42(H)	0.00424	-0.00086
43(H)	0.01288	-0.00007
44(H)	0.02985	-0.00001
45(H)	0.01884	-0.00002
46(H)	0.01686	-0.00001
47(H)	0.02067	-0.00003
48(H)	0.00261	-0.00135
49(H)	0.00091	-0.00159
50(H)	-0.0002	-0.00268
51(H)	0.00004	-0.00258
52(H)	0.00069	-0.00159
53(H)	0.00247	-0.00108
54(H)	0.00031	-0.00108
55(H)	-0.00008	-0.00007
56(H)	-0.00016	-0.00006
57(H)	0.00369	-0.00008
58(H)	0.05769	0.00000
59(H)	0.03494	-0.00001

Table S5 Condensed local electrophilicity/nucleophilicity index of tetracycline intermediate (molecular-3).

Atom	Electrophilicity	Nucleophilicity
1(C)	0.16036	0.01459
2(C)	0.08239	0.00973

3(C)	0.11050	0.01489
4(C)	0.06367	0.00974
5(C)	0.09460	0.00370
6(C)	0.06624	0.01316
7(C)	0.32182	0.01862
8(C)	0.07071	0.09833
9(C)	0.01364	0.01396
10(C)	0.01350	0.00431
11(C)	0.20008	0.05535
12(C)	0.02485	0.00410
13(C)	0.01008	0.00908
14(C)	0.01025	0.01612
15(C)	0.00683	0.01191
16(C)	0.00766	0.03835
17(C)	0.00804	0.01561
18(C)	0.00262	0.02531
19(O)	0.08887	0.01759
20(O)	0.03356	0.00755
21(C)	0.01309	0.00436
22(O)	0.33280	0.03838
23(O)	0.11961	0.07618
24(N)	0.0033	0.25032
25(C)	0.00555	0.06004
26(O)	0.01027	0.02287
27(C)	0.00419	0.01146
28(O)	0.00716	0.01815
29(N)	0.00358	0.00991

30(O)	0.02061	0.03214
31(O)	0.03966	0.01879
32(H)	0.06201	0.00622
33(H)	0.04189	0.00561
34(H)	0.03474	0.00596
35(H)	0.02722	0.02741
36(H)	0.02413	0.02233
37(H)	0.01504	0.01725
38(H)	0.01921	0.01844
39(H)	0.00965	0.02975
40(H)	0.04629	0.00687
41(H)	0.02038	0.00587
42(H)	0.01498	0.00465
43(H)	0.01167	0.00382
44(H)	0.01360	0.00537
45(H)	0.04208	0.02325
46(H)	0.00631	0.06215
47(H)	0.00363	0.04243
48(H)	0.00439	0.03668
49(H)	0.00448	0.07204
50(H)	0.00456	0.01219
51(H)	0.00282	0.00705
52(H)	0.00271	0.00508
53(H)	0.02955	0.01218

Table S6 Condensed local electrophilicity/nucleophilicity index of tetracycline intermediate (molecular-4).

Atom	Electrophilicity	Nucleophilicity
1(C)	0.13682	0.00545
2(C)	0.12743	0.00393
3(C)	0.06381	0.00428
4(C)	0.11682	0.00162
5(C)	0.13498	0.00297
6(C)	0.07499	0.00341
7(C)	0.10719	0.01409
8(C)	0.14496	-0.00233
9(C)	0.02832	0.00669
10(C)	0.05835	0.00366
11(C)	0.02920	0.02191
12(C)	0.00791	0.12977
13(C)	0.00949	0.04623
14(C)	0.02463	0.02107
15(C)	0.00323	0.14583
16(C)	0.00262	0.37639
17(C)	0.00610	0.08032
18(C)	0.00243	0.19596
19(O)	0.03591	0.0029
20(O)	0.01419	0.01408
21(C)	0.14727	0.01244
22(O)	0.04032	0.00586
23(O)	0.03814	0.01874
24(N)	0.00242	0.04082
25(O)	0.00469	0.08021

26(C)	0.00246	0.09009
27(O)	0.00381	0.15101
28(N)	0.00187	0.08484
29(O)	0.00708	0.3188
30(O)	0.01352	0.07564
31(O)	0.06227	0.00774
32(H)	0.05594	0.00245
33(H)	0.05360	0.00217
34(H)	0.03966	0.00211
35(H)	0.04645	0.01365
36(H)	0.02379	0.02909
37(H)	0.00275	0.07859
38(H)	0.01957	0.01805
39(H)	0.02237	0.02824
40(H)	0.00187	0.04073
41(H)	0.02044	0.00138
42(H)	0.01338	0.00565
43(H)	0.05265	0.01404
44(H)	0.05176	0.00582
45(H)	0.00635	0.01049
46(H)	0.01970	0.01307
47(H)	0.00216	0.03650
48(H)	0.00258	0.02865
49(H)	0.00199	0.04090
50(H)	0.00140	0.04713
51(H)	0.00112	0.03400
52(H)	0.00825	0.03012
53(H)	0.02263	0.00263

Table S7 Condensed local electrophilicity/nucleophilicity index of tetracycline intermediate (molecular-5).

Atom	Electrophilicity	Nucleophilicity
1(C)	0.16038	0.06057
2(C)	0.08318	0.05234
3(C)	0.10820	0.08549
4(C)	0.06589	0.07265
5(C)	0.09523	0.01992
6(C)	0.06587	0.09225
7(C)	0.30772	0.02359
8(C)	0.06336	0.16339
9(C)	0.01294	0.02030
10(C)	0.01363	0.00721
11(C)	0.19150	0.10935
12(C)	0.02260	0.00683
13(C)	0.00883	0.00506
14(C)	0.00942	0.01604
15(C)	0.00628	0.00361
16(C)	0.00733	0.00516
17(C)	0.00776	0.00471
18(C)	0.00367	0.00284
19(O)	0.08660	0.11328
20(O)	0.03229	0.01326
21(C)	0.01283	0.00843
22(O)	0.31666	0.04456
23(O)	0.12133	0.13289
24(N)	0.00587	0.00572

25(O)	0.00957	0.00628
26(C)	0.00405	0.00270
27(O)	0.00683	0.00459
28(N)	0.00341	0.00231
29(O)	0.01847	0.01091
30(O)	0.03808	0.01821
31(H)	0.06205	0.02636
32(H)	0.04184	0.02633
33(H)	0.03445	0.03115
34(H)	0.02626	0.03649
35(H)	0.02115	0.01377
36(H)	0.01443	0.01372
37(H)	0.01803	0.02066
38(H)	0.00996	0.00746
39(H)	0.04484	0.02813
40(H)	0.01968	0.01011
41(H)	0.01466	0.00939
42(H)	0.01140	0.00748
43(H)	0.01322	0.00955
44(H)	0.03891	0.03452
45(H)	0.00899	0.00597
46(H)	0.01120	0.00822
47(H)	0.00435	0.00290
48(H)	0.00267	0.00178
49(H)	0.00262	0.00171
50(H)	0.02891	0.01398

Table S8 Condensed local electrophilicity/nucleophilicity index of tetracycline intermediate (molecular-6).

Atom	Electrophilicity	Nucleophilicity
1(C)	0.00293	0.00167
2(C)	0.00237	0.00130
3(C)	0.00208	0.00127
4(C)	0.00083	0.00057
5(C)	0.00091	0.00062
6(C)	0.00224	0.00126
7(C)	0.00302	0.00185
8(C)	0.00258	0.00058
9(C)	0.00272	0.00136
10(C)	0.00284	0.00173
11(C)	0.01221	0.00626
12(C)	0.01319	0.00539
13(C)	0.03326	0.01340
14(C)	0.02164	0.01164
15(C)	0.65066	0.12623
16(C)	0.51170	0.08914
17(C)	0.07208	0.04176
18(O)	0.00146	0.00096
19(O)	0.00326	0.00181
20(O)	0.00587	0.00412
21(O)	0.01340	0.00774
22(N)	0.04898	0.03307
23(O)	0.62113	0.20257
24(O)	0.69649	0.24565
25(O)	0.01941	0.04205

26(O)	0.00922	0.00467
27(H)	0.00152	0.00082
28(H)	0.00136	0.00073
29(H)	0.00151	0.00083
30(H)	0.00715	0.00407
31(H)	0.01276	0.00772
32(H)	0.03959	0.01506
33(H)	0.01583	0.00737
34(H)	0.02255	0.01169
35(H)	0.17993	0.07226
36(H)	0.08521	0.02752
37(H)	0.00304	0.00161
38(H)	0.00802	0.00431
39(H)	0.04320	0.02236
40(H)	0.03957	0.01865
41(H)	0.01948	0.01458
42(C)	0.00219	0.00117
43(H)	0.00249	0.00124
44(H)	0.00147	0.00087
45(H)	0.00272	0.00138
46(H)	0.00303	0.00148
47(H)	0.00104	0.00061
48(H)	0.05135	0.01509

Table S9 Condensed local electrophilicity/nucleophilicity index of tetracycline intermediate (molecular-7).

Atom	Electrophilicity	Nucleophilicity
1(C)	0.00232	0.06631

2(C)	0.00210	0.06277
3(C)	0.00197	0.09836
4(C)	0.00086	0.08382
5(C)	0.00060	0.02401
6(C)	0.00179	0.10953
7(C)	0.00483	0.02467
8(C)	0.00648	0.15822
9(C)	0.00392	0.01832
10(C)	0.00199	0.00738
11(C)	0.00380	0.10841
12(C)	0.00632	0.01504
13(C)	0.01743	0.00791
14(C)	0.01408	0.01599
15(C)	0.54257	0.00202
16(C)	0.47637	0.00276
17(C)	0.06584	0.00349
18(O)	0.00220	0.13415
19(O)	0.00262	0.01402
20(C)	0.00231	0.00848
21(O)	0.00777	0.04306
22(O)	0.00785	0.12042
23(N)	0.07851	0.00555
24(O)	0.51009	0.00472
25(O)	0.58205	0.00339
26(O)	0.03176	0.01761
27(H)	0.00115	0.02956
28(H)	0.00117	0.03072

29(H)	0.00104	0.03619
30(H)	0.00712	0.03154
31(H)	0.01349	0.02602
32(H)	0.02653	0.01154
33(H)	0.01551	0.0180
34(H)	0.01683	0.01595
35(H)	0.20875	0.00161
36(H)	0.09813	0.00623
37(H)	0.00113	0.03184
38(H)	0.00233	0.01038
39(H)	0.00196	0.0079
40(H)	0.00255	0.00976
41(H)	0.00453	0.00928
42(H)	0.00567	0.03427
43(H)	0.07167	0.00385
44(H)	0.05435	0.00424
45(H)	0.04049	0.00953

Table S10 Condensed local electrophilicity/nucleophilicity index of tetracycline intermediate (molecular-8).

Atom	Electrophilicity	Nucleophilicity
1(C)	0.23169	0.05908
2(C)	0.11891	0.13581
3(C)	0.15177	0.12153
4(C)	0.10325	0.07505
5(C)	0.14233	0.07980

6(C)	0.08957	0.18690
7(C)	0.35587	0.02801
8(C)	0.03808	0.00435
9(C)	0.00926	0.00305
10(C)	0.01688	0.00833
11(C)	0.02090	0.00352
12(C)	0.00940	0.00175
13(C)	0.00699	0.00226
14(C)	0.01218	0.00401
15(O)	0.11625	0.16942
16(O)	0.03479	0.02214
17(C)	0.01524	0.00825
18(O)	0.36102	0.05216
19(O)	0.01018	0.00221
20(O)	0.06721	0.00878
21(H)	0.08922	0.03469
22(H)	0.05851	0.05131
23(H)	0.04583	0.05508
24(H)	0.02282	0.00639
25(H)	0.02654	0.00376
26(H)	0.02300	0.00416
27(H)	0.00906	0.00182
28(H)	0.00873	0.00209
29(H)	0.01046	0.00291
30(H)	0.01247	0.00406
31(H)	0.01377	0.00396
32(H)	0.05329	0.03975

33(H)	0.02152	0.01188
34(H)	0.01477	0.00928
35(H)	0.01453	0.00831
36(H)	0.01703	0.01072
37(H)	0.00737	0.00151
38(H)	0.03424	0.00536

Table S11 Condensed local electrophilicity/nucleophilicity index of tetracycline intermediate (molecular-9).

Atom	Electrophilicity	Nucleophilicity
1(C)	0.02974	0.02517
2(C)	0.05073	0.05522
3(C)	0.24198	0.09077
4(C)	0.04330	0.04592
5(C)	0.00703	0.03516
6(C)	0.02740	0.03942
7(C)	0.12598	0.00575
8(C)	0.03814	0.00701
9(C)	0.00417	0.00718
10(C)	0.00942	0.01615
11(O)	0.02704	0.00887
12(C)	0.00305	0.00532
13(O)	0.17149	0.03641
14(N)	0.00180	0.00309
15(O)	0.27691	0.28386
16(C)	0.01300	0.01386

17(O)	0.06382	0.07702
18(H)	0.02614	0.02295
19(H)	0.03196	0.03357
20(H)	0.03969	0.03873
21(H)	0.06067	0.03874
22(H)	0.05348	0.01149
23(H)	0.03149	0.00729
24(H)	0.00588	0.00649
25(H)	0.00687	0.00615
26(H)	0.01473	0.01553
27(H)	0.01097	0.01248
28(H)	0.01903	0.00483
29(H)	0.00307	0.00385
30(H)	0.00239	0.00337
31(H)	0.00169	0.00268
32(H)	0.00123	0.00171
33(H)	0.01072	0.01178
34(H)	0.01141	0.01242
35(H)	0.00852	0.00899
36(H)	0.03736	0.02622

Table S12 Condensed local electrophilicity/nucleophilicity index of tetracycline intermediate (molecular-10).

Atom	Electrophilicity	Nucleophilicity
1(C)	0.14651	0.1082

2(C)	0.13347	0.15877
3(C)	0.05284	0.19203
4(C)	0.09874	0.1148
5(C)	0.09830	0.07601
6(C)	0.06093	0.25594
7(C)	0.01932	0.02081
8(C)	0.00623	0.00908
9(C)	0.01112	0.01078
10(C)	0.02363	0.02190
11(C)	0.00347	0.00541
12(O)	0.00538	0.00663
13(O)	0.03093	0.24651
14(O)	0.01125	0.01778
15(O)	0.00887	0.01089
16(H)	0.06034	0.05319
17(H)	0.05604	0.06609
18(H)	0.03129	0.08219
19(H)	0.02879	0.03373
20(H)	0.01173	0.01113
21(H)	0.01424	0.01488
22(H)	0.01831	0.02215
23(H)	0.02730	0.02484
24(H)	0.00425	0.00608
25(H)	0.00568	0.00835
26(H)	0.00451	0.00550
27(H)	0.01183	0.0563
28(H)	0.01134	0.01711

29(H)	0.00700	0.00835

Table S13 Condensed local electrophilicity/nucleophilicity index of tetracycline intermediate (molecular-11).

Atom	Electrophilicity	Nucleophilicity
1(C)	0.15868	0.06212
2(C)	0.07521	0.05570
3(C)	0.10366	0.08949
4(C)	0.07622	0.07869
5(C)	0.09281	0.02367
6(C)	0.06415	0.09987
7(C)	0.28696	0.02255
8(C)	0.06196	0.13263
9(C)	0.01394	0.01765
10(C)	0.01039	0.00893
11(C)	0.21347	0.12066
12(O)	0.10813	0.10962
13(C)	0.01335	0.0108
14(O)	0.06262	0.10846
15(O)	0.02349	0.01670
16(C)	0.01620	0.00770
17(O)	0.31609	0.04650
18(H)	0.06117	0.02729
19(H)	0.03778	0.02689
20(H)	0.03432	0.03387
21(H)	0.02464	0.03535
22(H)	0.08028	0.04063

23(H)	0.04355	0.03677
24(H)	0.01183	0.01081
25(H)	0.01317	0.01049
26(H)	0.01452	0.01212
27(H)	0.02989	0.0346
28(H)	0.01639	0.0099
29(H)	0.01555	0.00949
30(H)	0.01803	0.00830
31(H)	0.01601	0.00858

Table S14 Condensed local electrophilicity/nucleophilicity index of tetracycline intermediate (molecular-12).

Atom	Electrophilicity	Nucleophilicity
1(C)	0.18944	-0.01109
2(C)	0.08037	-0.02419
3(C)	0.19627	-0.04154
4(O)	0.09848	-0.01115
5(O)	0.09897	-0.04349
6(O)	0.03779	-0.05555
7(O)	0.18384	-0.02949
8(O)	0.19093	-0.13937
9(H)	0.13234	-0.01855
10(H)	0.04294	-0.00625
11(H)	0.04408	-0.01818
12(H)	0.02534	-0.01661

Table S15 Condensed local electrophilicity/nucleophilicity index of tetracycline intermediate (molecular-13).

Atom	Electrophilicity	Nucleophilicity
1(C)	0.32306	-0.12824
2(C)	0.07412	-0.05519
3(C)	0.03749	-0.01630
4(O)	0.15131	-0.13432
5(O)	0.02658	-0.01626
6(O)	0.31215	-0.49484
7(O)	0.04026	-0.02155
8(H)	0.07400	-0.04039
9(H)	0.04572	-0.03126
10(H)	0.06571	-0.05224
11(H)	0.01810	-0.01078

Table S16 Condensed local electrophilicity/nucleophilicity index of tetracycline intermediate (molecular-14).

Atom	Electrophilicity	Nucleophilicity
1(C)	0.42931	-0.09345
2(C)	0.42931	-0.09345
3(O)	0.23149	-0.08974
4(O)	0.23149	-0.08974
5(O)	0.42472	-0.25222
6(O)	0.42472	-0.25222
7(H)	0.08845	-0.03404
8(H)	0.08845	-0.03404

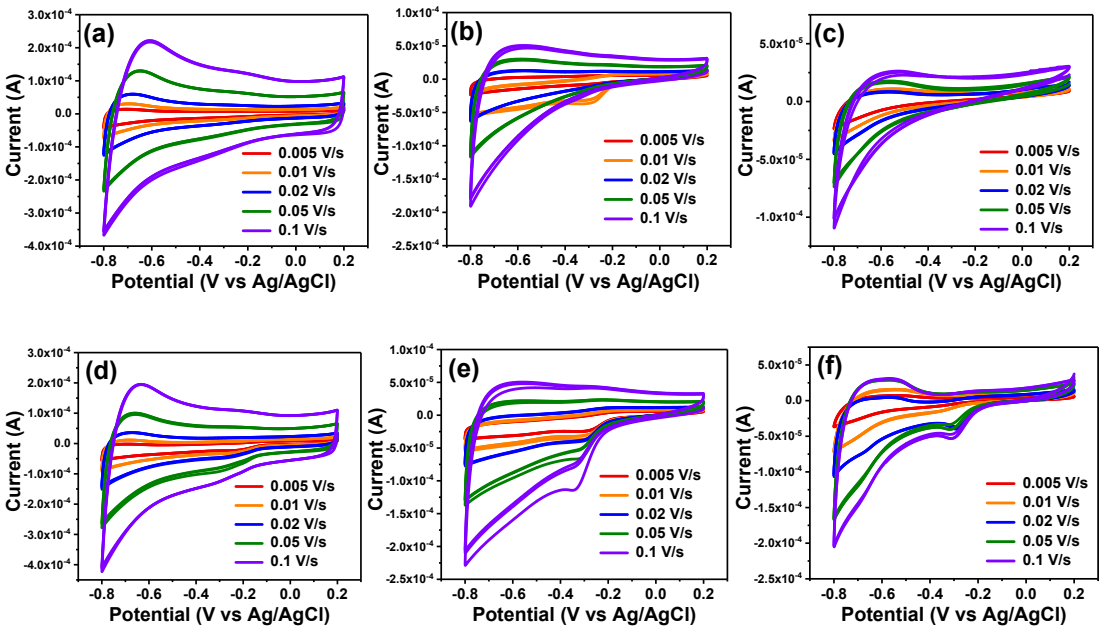


Figure S7 (a) Cyclic voltammograms of the ORR on AMG obtained in N_2-saturated 0.1 M KOH at 0.01 V/s, (b) cyclic voltammograms of the ORR on PMG obtained in N_2-saturated 0.1 M KOH at 0.01 V/s, (c) cyclic voltammograms of the ORR on DMG obtained in N_2-saturated 0.1 M KOH at 0.01 V/s, (d) Cyclic voltammograms of the ORR on AMG obtained in O_2-saturated 0.1 M KOH at 0.01 V/s, (e) cyclic voltammograms of the ORR on PMG obtained in O_2-saturated 0.1 M KOH at 0.01 V/s, (f) cyclic voltammograms of the ORR on DMG obtained in O_2-saturated 0.1 M KOH at 0.01 V/s.

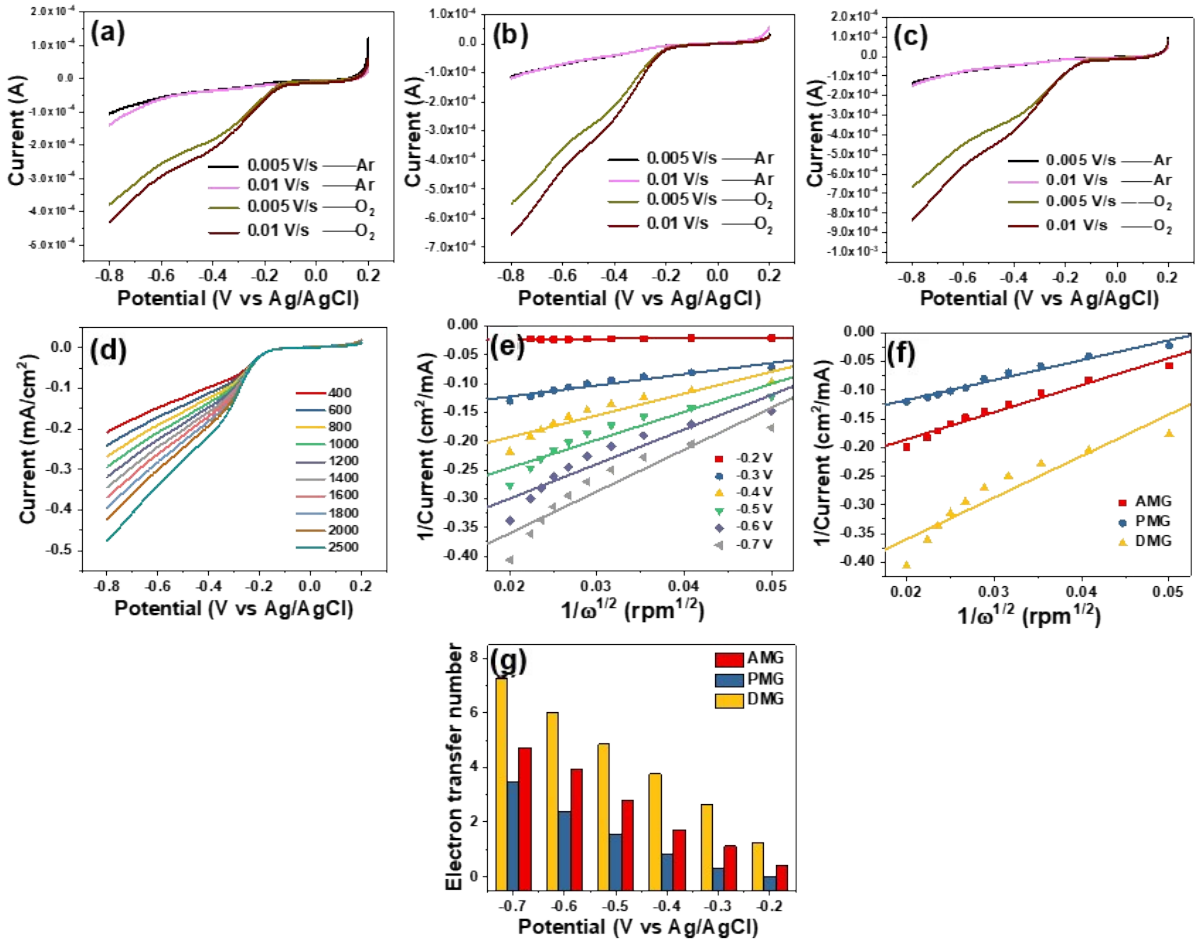


Figure S8 (a) RDE linear sweep voltammograms of the AMG in 0.1-M KOH, (b) RDE linear sweep voltammograms of the PMG in 0.1-M KOH, (c) RDE linear sweep voltammograms of the DMG in 0.1-M KOH , (d) rotating disk electrode (RDE) linear sweep voltammograms of DMG in O_2-saturated 0.1 M KOH with various rotation rates (scan rate 10 mV/s), (e) Koutecky−Levich plots of DMG under different voltammograms, (f) Koutecky−Levich plots of AMG, PMG and DMG, (g) electrochemical activity given as the full diffusion-limited current density.

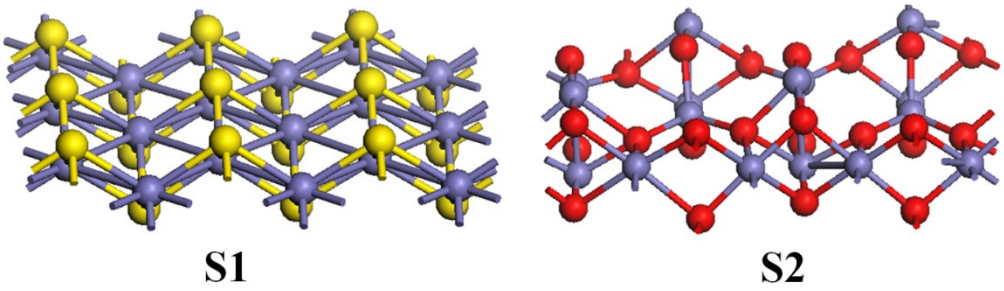


Figure S9 Computational models. Fe, O and S are shown as slateblue, red and yellow colours.