Supporting Information for

Temporally Controlled Supramolecular Catalysts with pH-Dependent Activity

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≠ these authors equally contributed to the present work.

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Section S1 Potentiometric Experiments

In the following pages are reported the plots and the experimental details of the potentiometric experiments carried out in the absence of any catalysts. Initial pH is adjusted with small aliquots of semi-aqueous $HClO_4$ 1.0 M, unless otherwise specified. Further details about the experiments and the definition of the DS duration are reported in the main text and in the Experimental Section of the article. In these experiments the pH read on the instrument is reported. In order to convert it to the real pH see the equation in the Experimental Section.

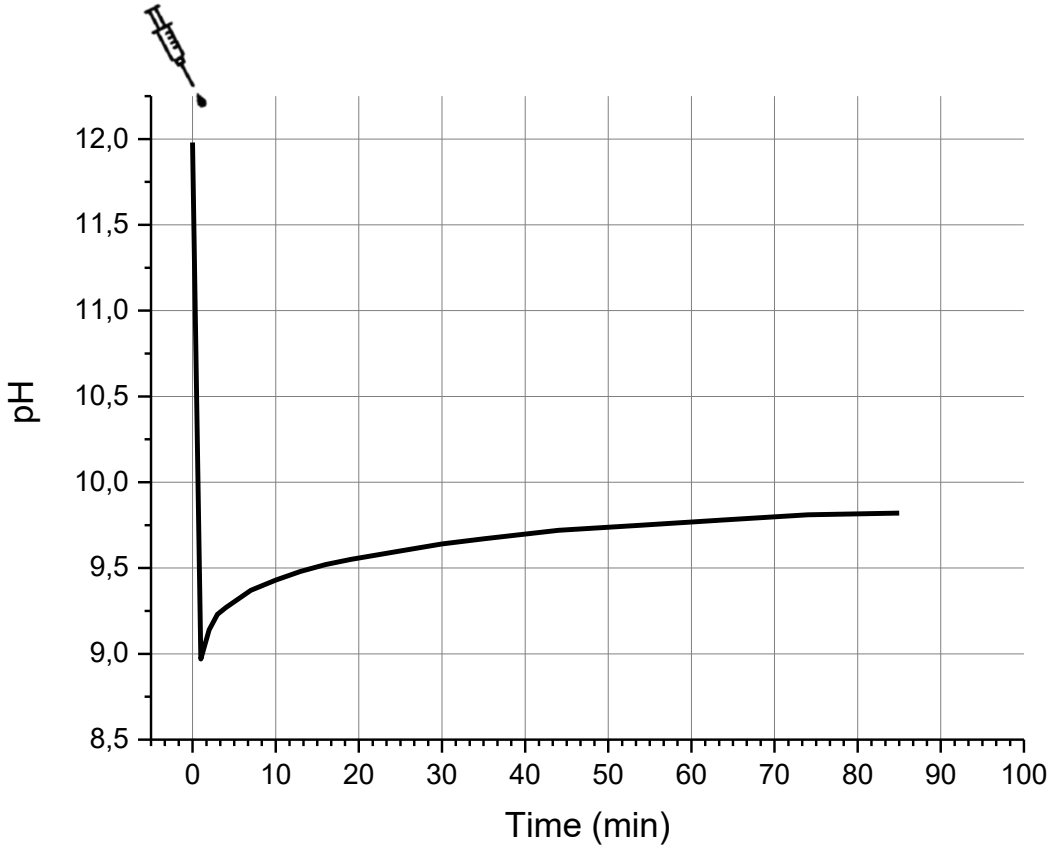


Figure S1.1

TCA inj. #	Initial pH	pH after TCA	pH plateau	DS duration (min)	[TCA] (mM)	[K_2CO_3] (mM)
1	11.98	8.97	9.82	n/a	5	10

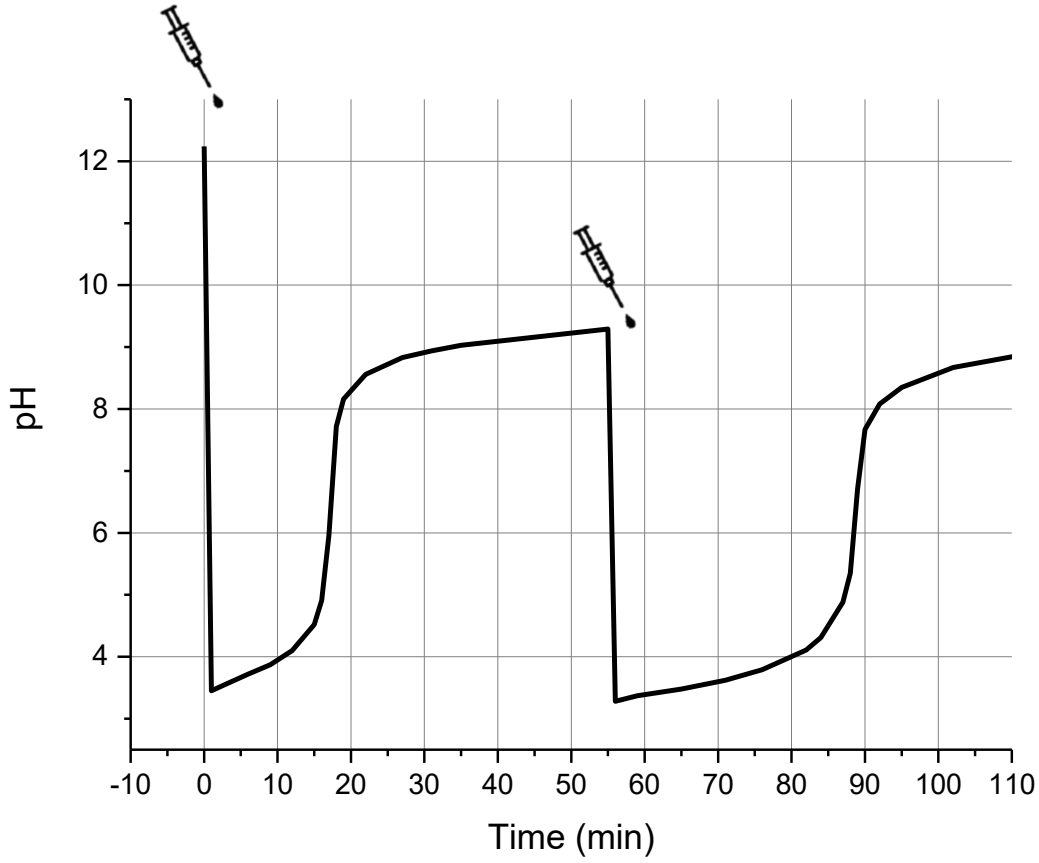


Figure S1.2

TCA inj. #	Initial pH	pH after TCA	pH plateau	DS duration (min)	[TCA] (mM)	[K_2CO_3] (mM)
1	12.24	3.45	9.29	14	7.5	10
2	9.29	3.28	8.95	30	7.5	10

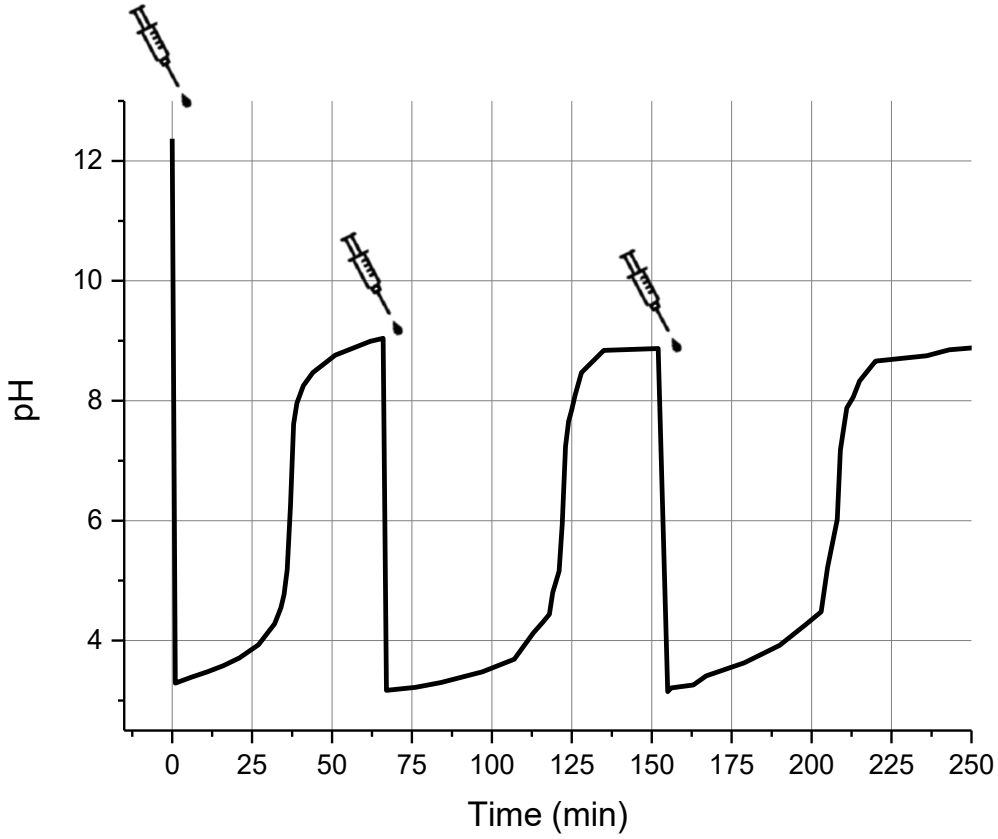


Figure S1.3

TCA inj. #	Initial pH	pH after TCA	pH plateau	DS duration (min)	[TCA] (mM)	[K_2CO_3] (mM)
1	12.37	3.29	9.04	33	10	10
2	9.04	3.17	8.87	50	10	10
3	8.87	3.15	8.9	48	10	10

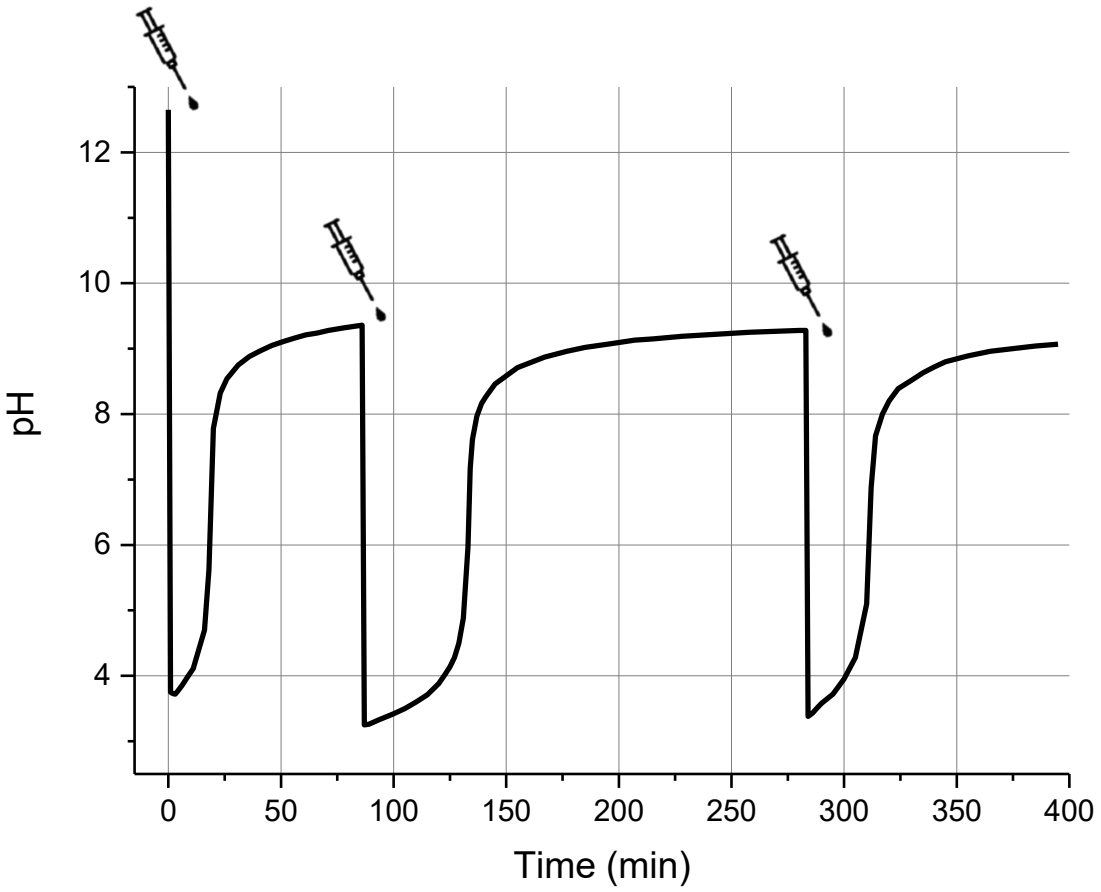


Figure S1.4

TCA inj. #	Initial pH	pH after TCA	pH plateau	DS duration (min)	[TCA] (mM)	[K_2CO_3] (mM)
1	12.65	3.72	9.36	15	10	10
2	9.36	3.25	9.28	43	10	10
3	9.28	3.38	9.07	26	10	10

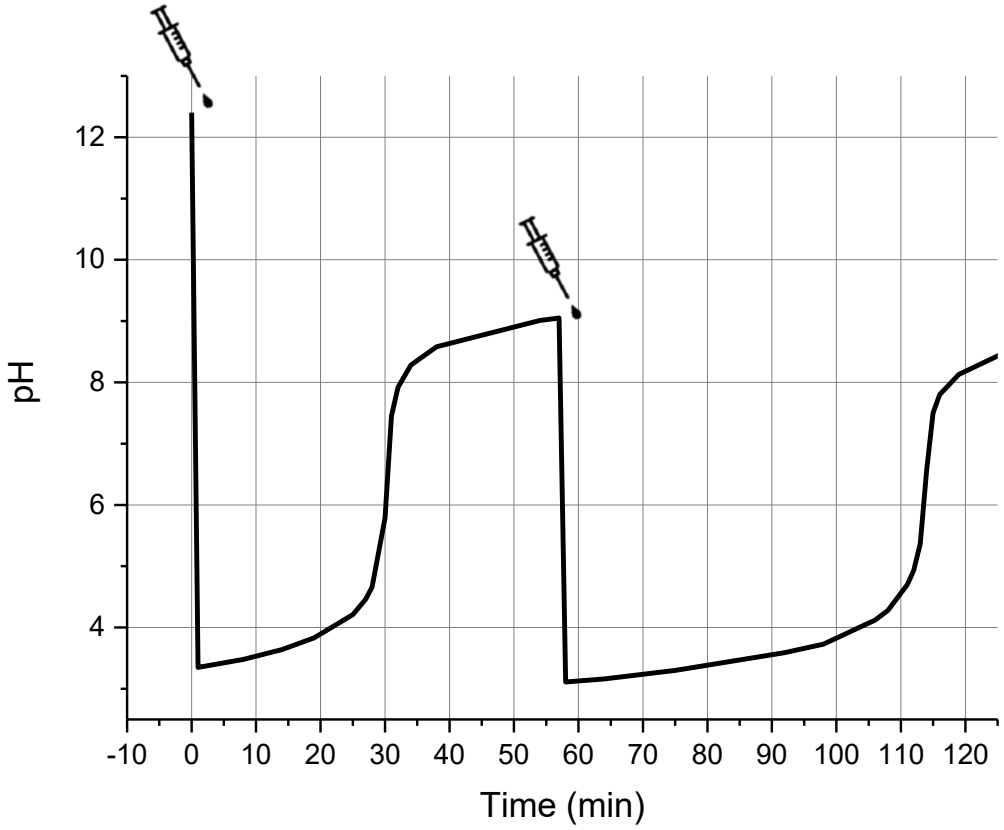


Figure S1.5

TCA inj. #	Initial pH	pH after TCA	pH plateau	DS duration (min)	[TCA] (mM)	[K_2CO_3] (mM)
1	12.4	3.35	9.05	27	15	10
2	9.05	3.11	8.66	52	15	10

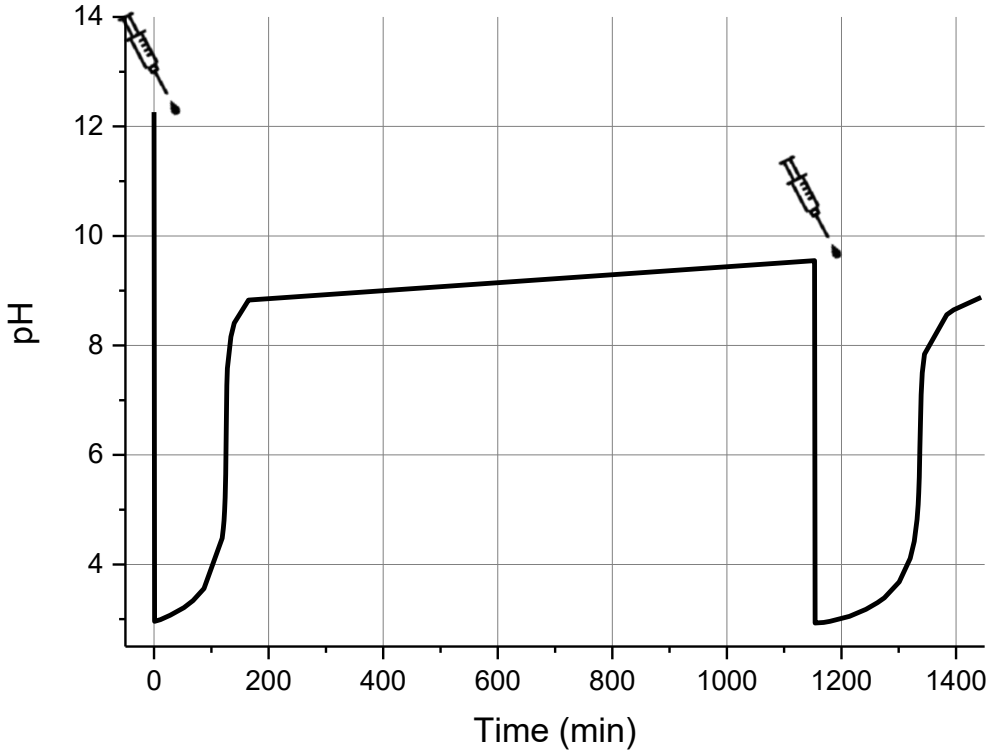


Figure S1.6

TCA inj. #	Initial pH	pH after TCA	pH plateau	DS duration (min)	[TCA] (mM)	[K_2CO_3] (mM)
1	12.26	2.96	9.55	124	20	10
2	9.55	2.93	8.65	182	20	10

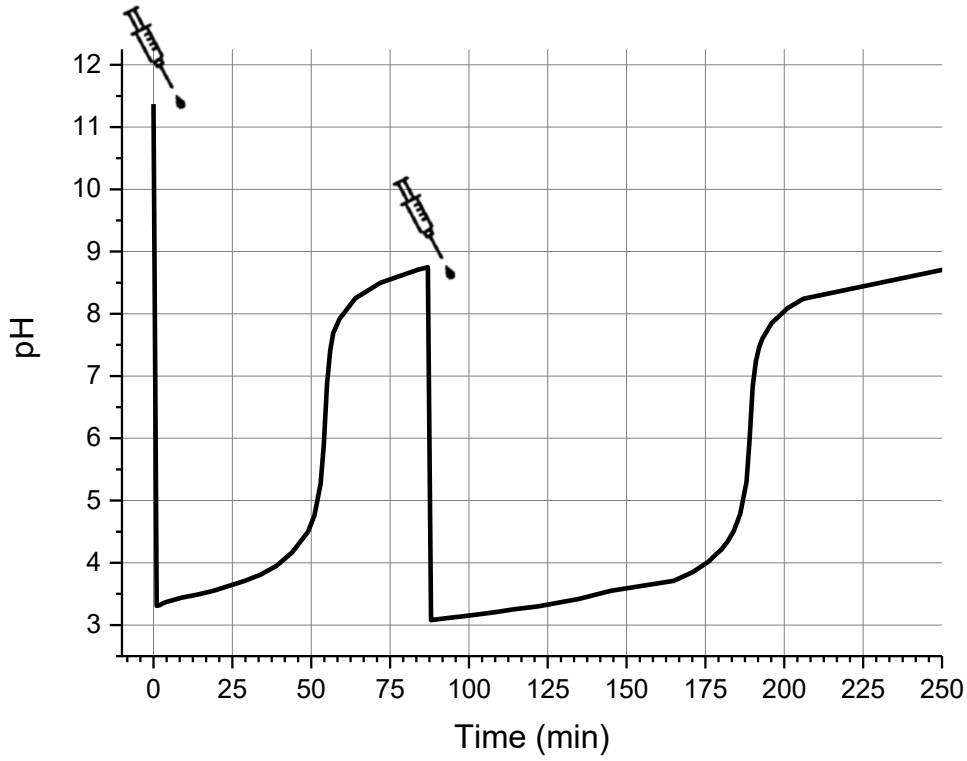


Figure S1.7

TCA inj. #	Initial pH	pH after TCA	pH plateau	DS duration (min)	[TCA] (mM)	[K_2CO_3] (mM)
1	11.37	3.31	8.75	50	10	10
2	8.75	3.08	8.77	97	10	10

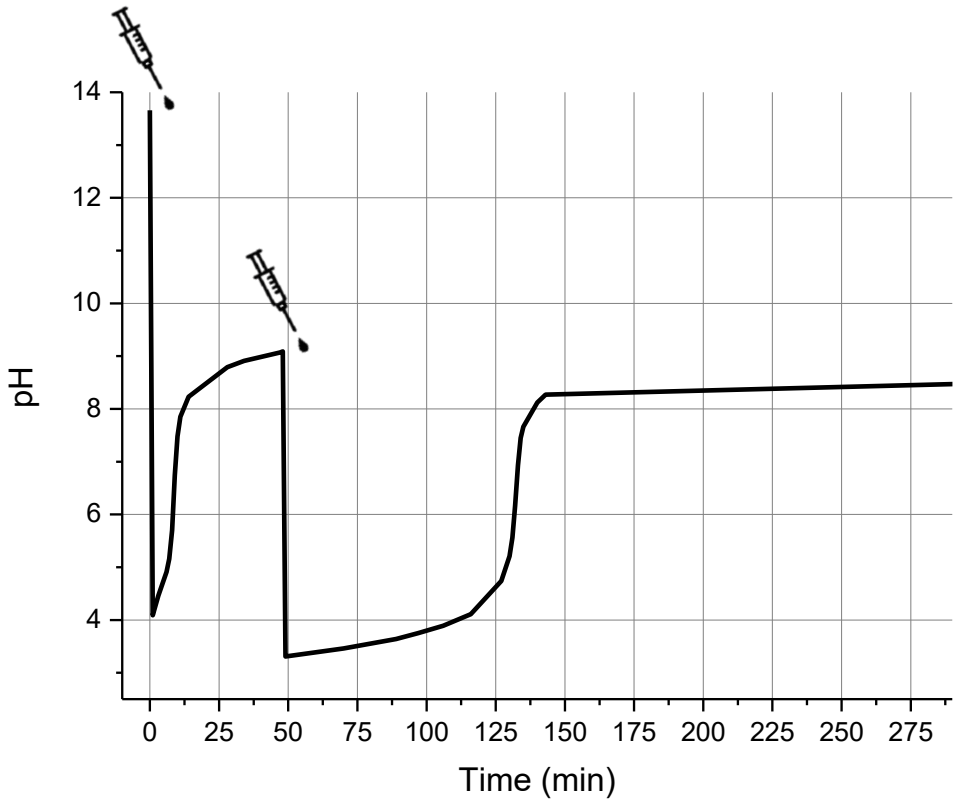


Figure S1.8

TCA inj. #	Initial pH	pH after TCA	pH plateau	DS duration (min)	[TCA] (mM)	[K_2CO_3] (mM)
1	13.66	4.09	9.09	7	10	10
2	9.09	3.31	9.49	81	10	10

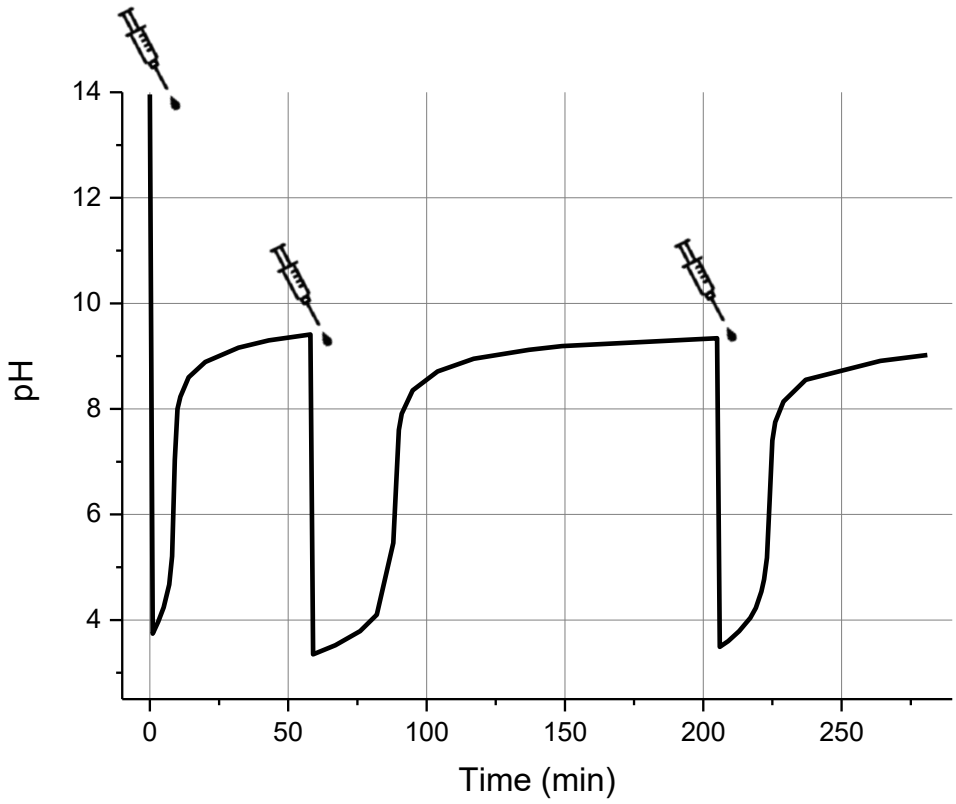


Figure S1.9

TCA inj. #	Initial pH	pH after TCA	pH plateau	DS duration (min)	[TCA] (mM)	[K_2CO_3] (mM)
1	13.96	3.74	9.41	8	10	10
2	9.41	3.35	9.34	31	10	10
3	9.34	3.35	9.12	20	10	10

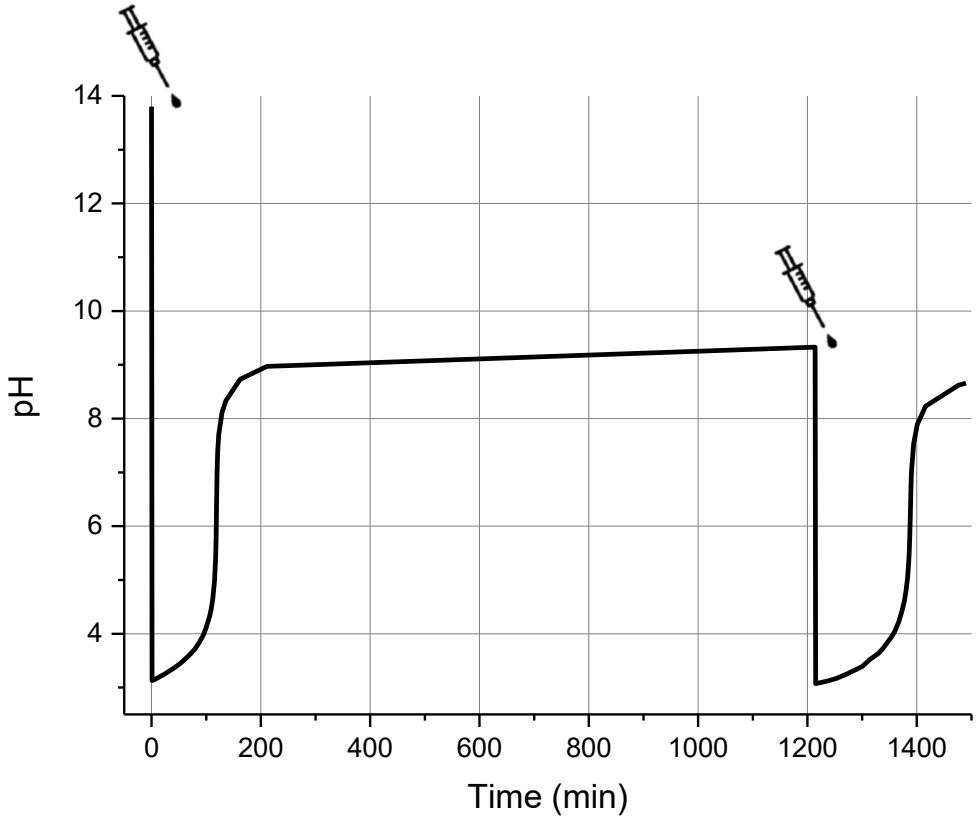


Figure S1.10

TCA inj. #	Initial pH	pH after TCA	pH plateau	DS duration (min)	[TCA] (mM)	[K_2CO_3] (mM)
1	13.8	3.13	9.33	115	10	5
2	9.33	3.07	8.66	168	10	5

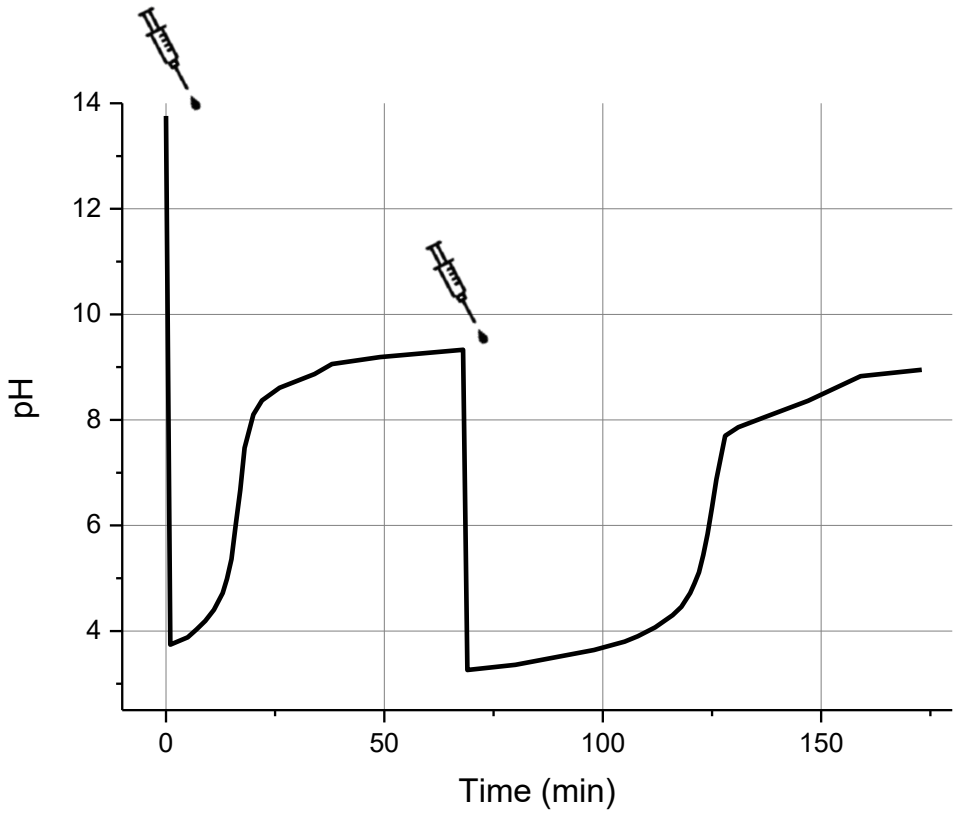


Figure S1.11

TCA inj. #	Initial pH	pH after TCA	pH plateau	DS duration (min)	[TCA] (mM)	[K_2CO_3] (mM)
1	13.76	3.74	9.33	13	10	15
2	9.33	3.26	8.95	54	10	15

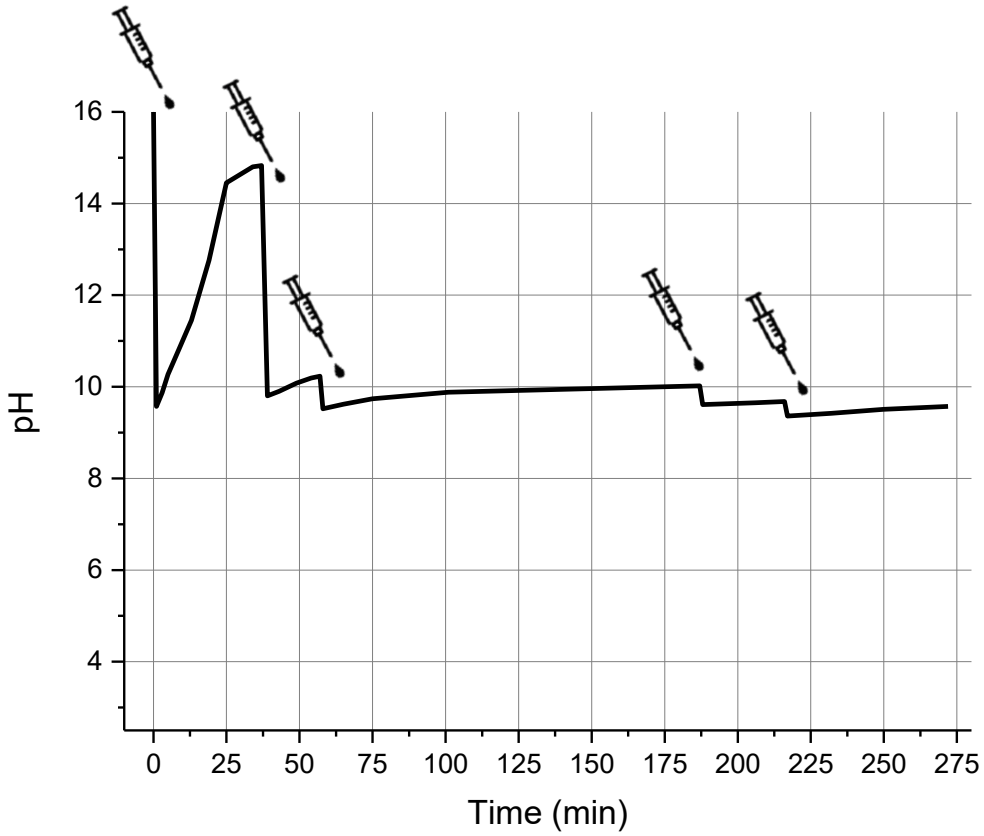


Figure S1.12

TCA inj. #	Initial pH	pH after TCA	pH plateau	DS duration (min)	[TCA] (mM)	[K_2CO_3] (mM)
1	16 [a]	9.57	14.83	n/a	5	10
2	14.83	9.8	10.23	n/a	5	10
3	10.23	9.52	10.02	n/a	5	10
4	10.02	9.61	9.68	n/a	5	10
5	9.68	9.36	9.57	n/a	5	10

[a] No pH pre-adjustment.

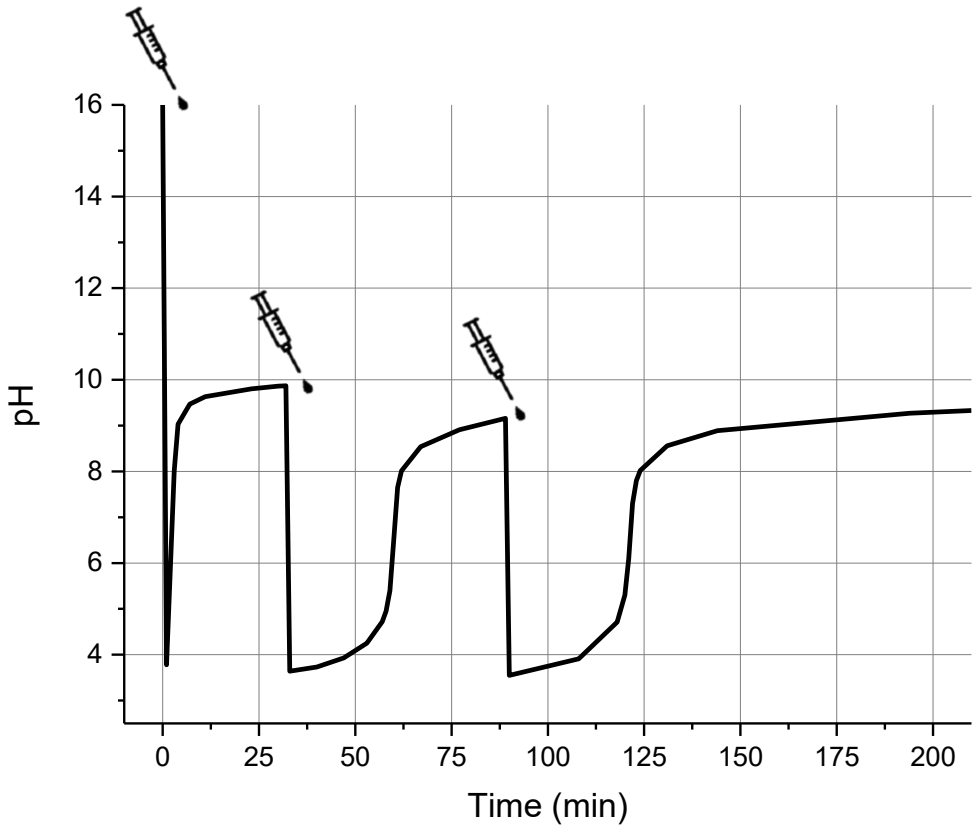


Figure S1.13

TCA inj. #	Initial pH	pH after TCA	pH plateau	DS duration (min)	[TCA] (mM)	[K_2CO_3] (mM)
1	16	3.78	9.87	1	7.5	10
2	9.87	3.64	9.16	26	7.5	10
3	9.16	3.55	9.33	30	7.5	10

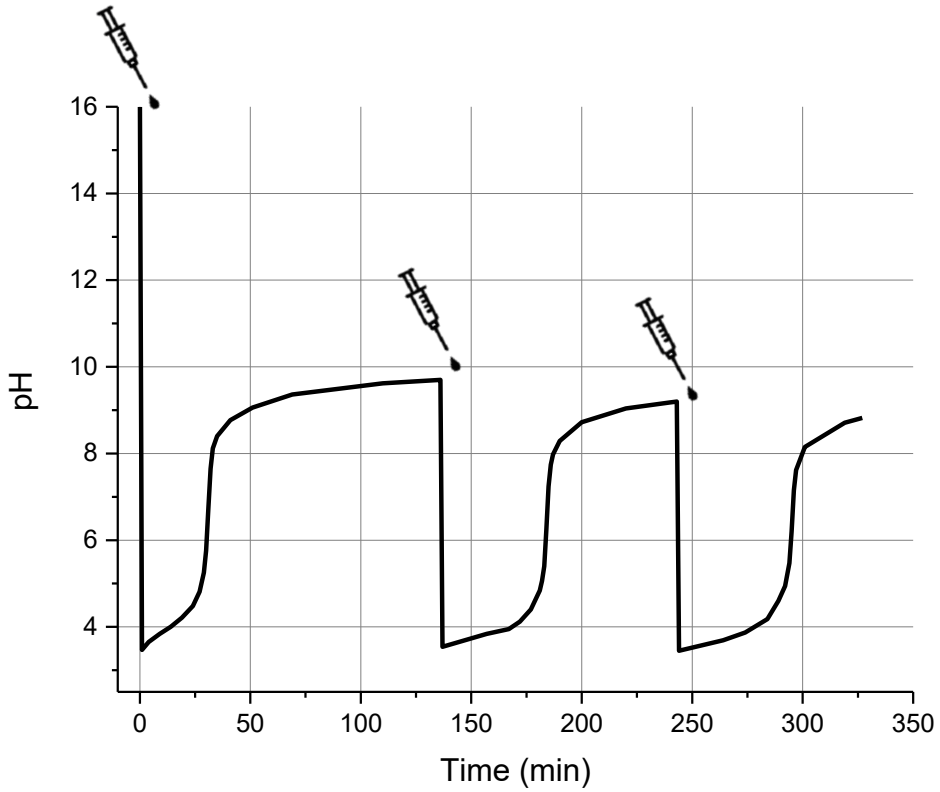


Figure S1.14

TCA inj. #	Initial pH	pH after TCA	pH plateau	DS duration (min)	[TCA] (mM)	[K_2CO_3] (mM)
1	16	3.47	9.7	29	10	10
2	9.7	3.54	9.2	47	10	10
3	9.2	3.45	9.39	51	10	10

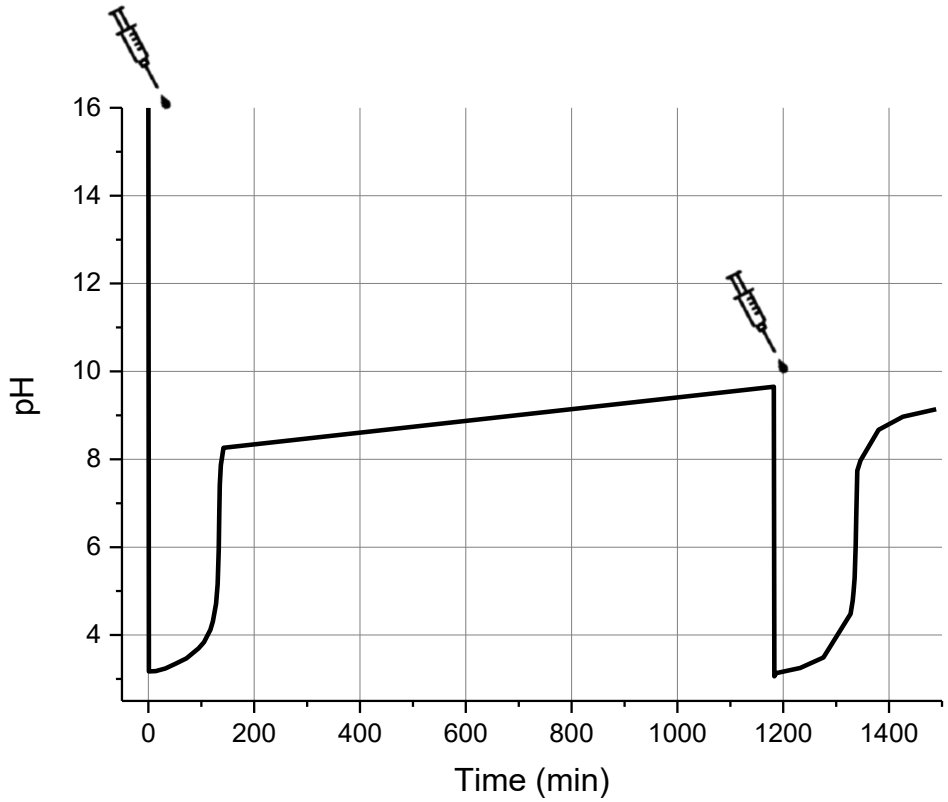


Figure S1.15

TCA inj. #	Initial pH	pH after TCA	pH plateau	DS duration (min)	[TCA] (mM)	[K_2CO_3] (mM)
1	16	3.17	9.65	131	20	10
2	9.65	3.06	9.14	153	20	10

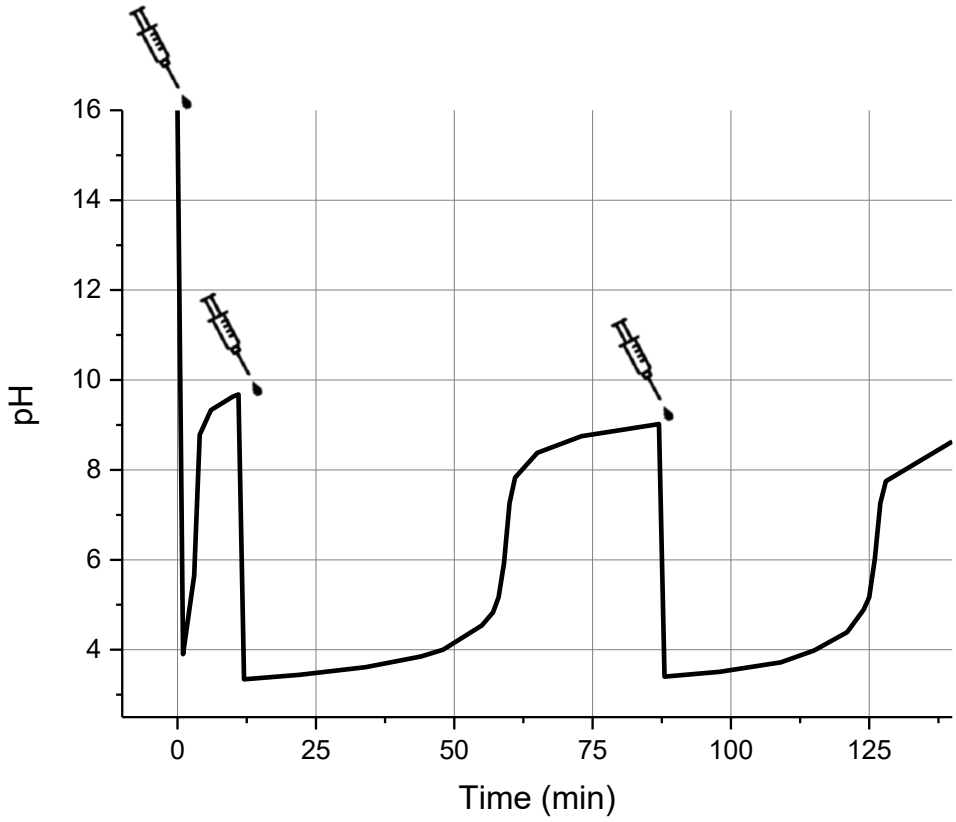


Figure S1.16

TCA inj. #	Initial pH	pH after TCA	pH plateau	DS duration (min)	[TCA] (mM)	[K_2CO_3] (mM)
1	16	3.9	9.68	2	7.5	10
2	9.68	3.34	9.02	46	10	10
3	9.02	3.4	8.63	38	8.75	10

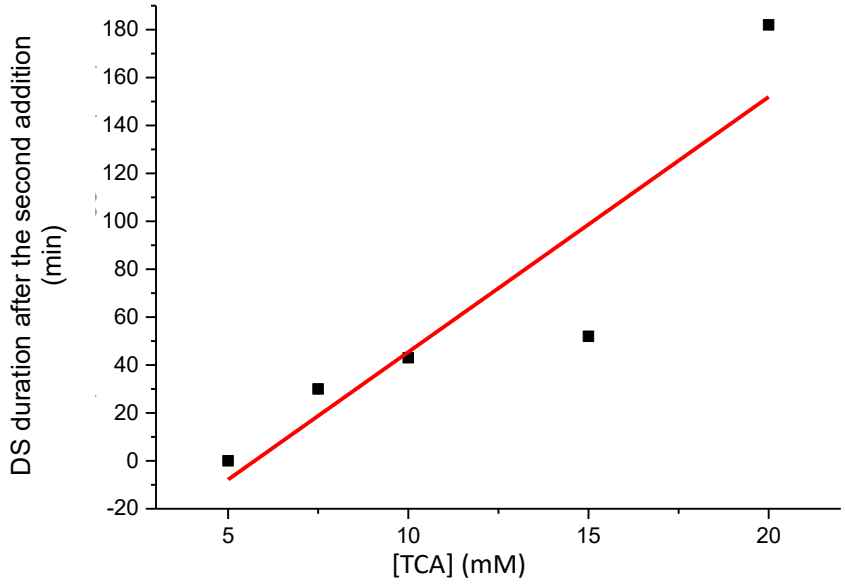


Figure S1.17 Plot of the duration of the dissipative state after the second addition of TCA versus the TCA concentration. In the corresponding experiments [K_2CO_3] = 10 mM The solution was pre-adjusted with $HClO_4$ at an approximate value of/ at about pH 12.5.

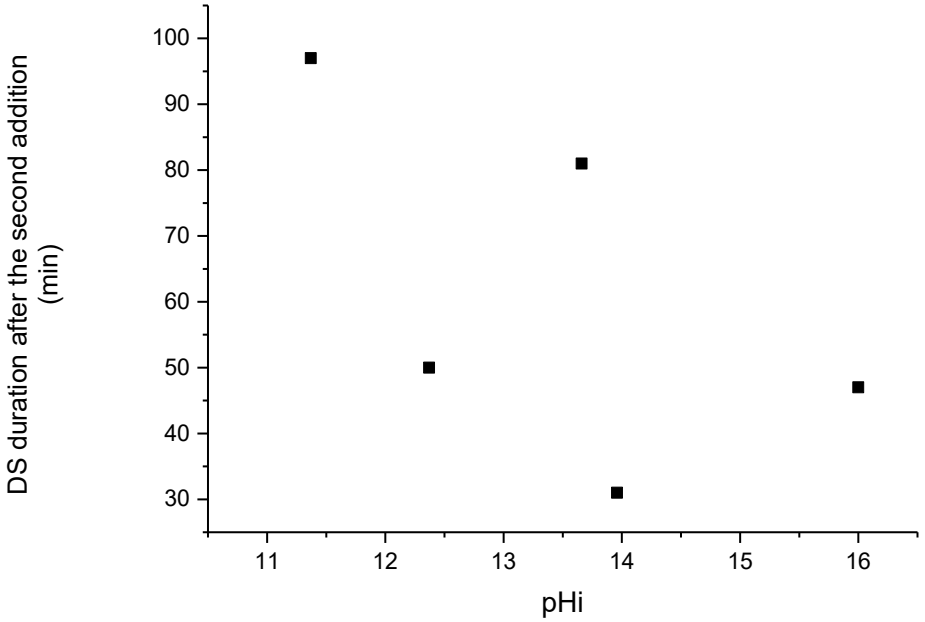


Figure S1.18 Plot of the duration of the dissipative state after the second addition of TCA versus the initial pH pre-adjusted with $HClO_4$ / the plateau pH at the end of the first dissipative cycle. [K_2CO_3] = 10 mM and [TCA] = 10 mM.

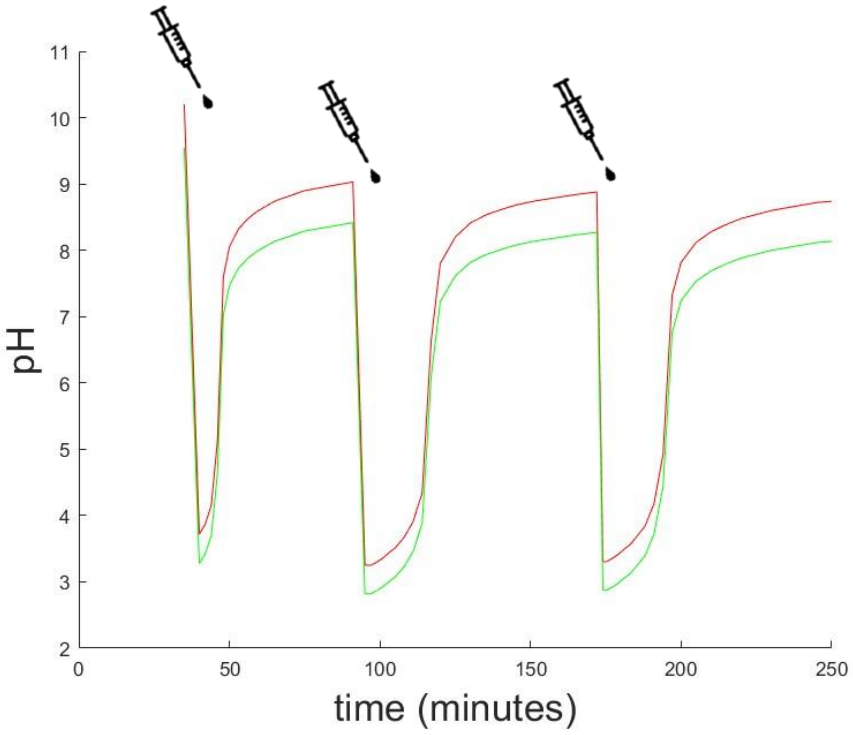


Figure S1.19 Plot of uncorrected (red) and corrected (green) pH. The pH values reported in the table below are corrected ones.

TCA inj. #	Initial pH	pH after TCA	pH plateau	DS duration (min)	[TCA] (mM)	[K_2HPO_4] (mM)
1	10.20	*3.72*	9.03	8	7.5	10
2	9.03	3.25	8.88	22	10	10
3	8.88	3.31	8.74	22	8.75	10

Section S2 Decarboxylation of TCA monitored with UV-Vis Spectrophotometry

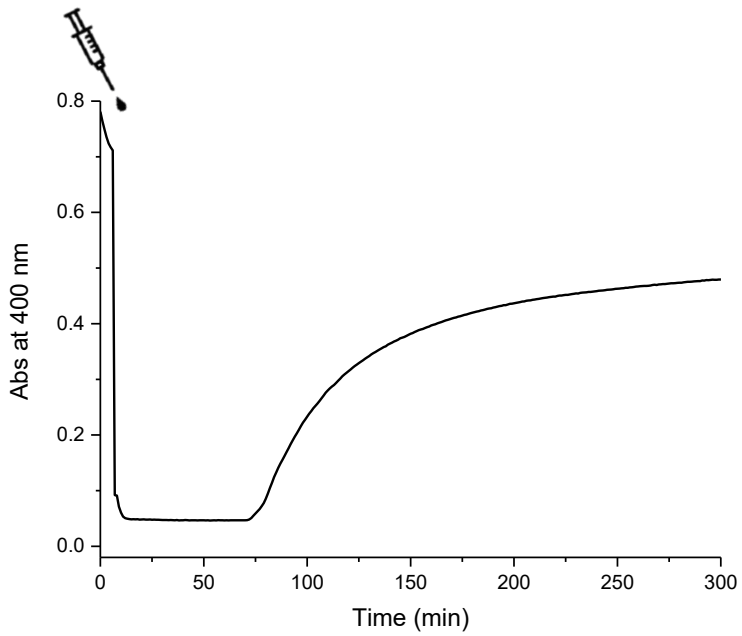


Figure S2.1 Plot of the absorbance at 400 nm versus time for a $5.0 \cdot 10^{-5}$ M *p*-nitrophenol solution in 80% DMSO, 10 mM K_2CO_3, upon an addition of 20.0 mM of trichloroacetic acid. This experiment provided the molar extinction coefficient for *p*-nitrophenol and *p*-nitrophenolate reported in the Experimental Section.

Section S3 Kinetic Experiments in the presence of the Catalysts

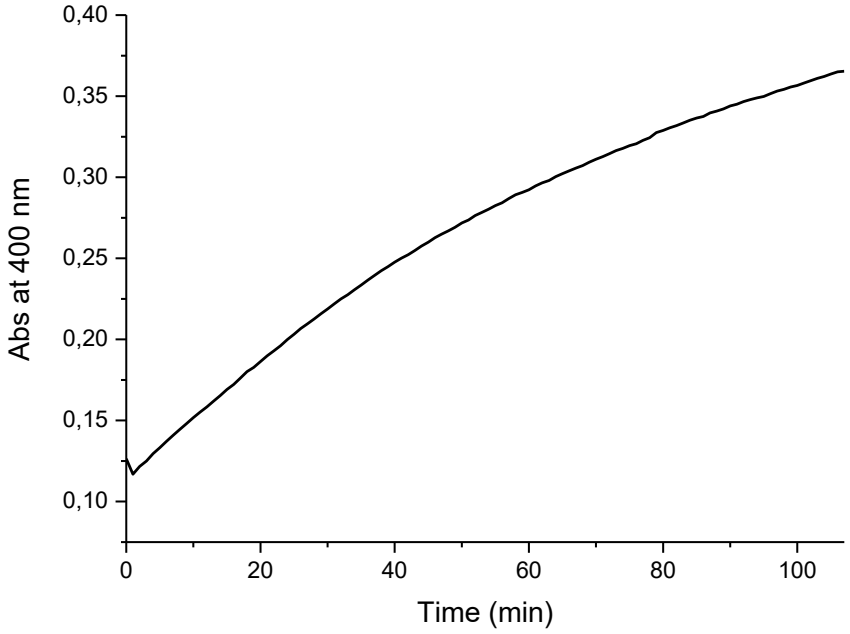


Figure S3.1 **1**-Cu^{II} 2.0 mM, HPNP 0.2 mM, TCA 10 mM, no prior pH adjustment.

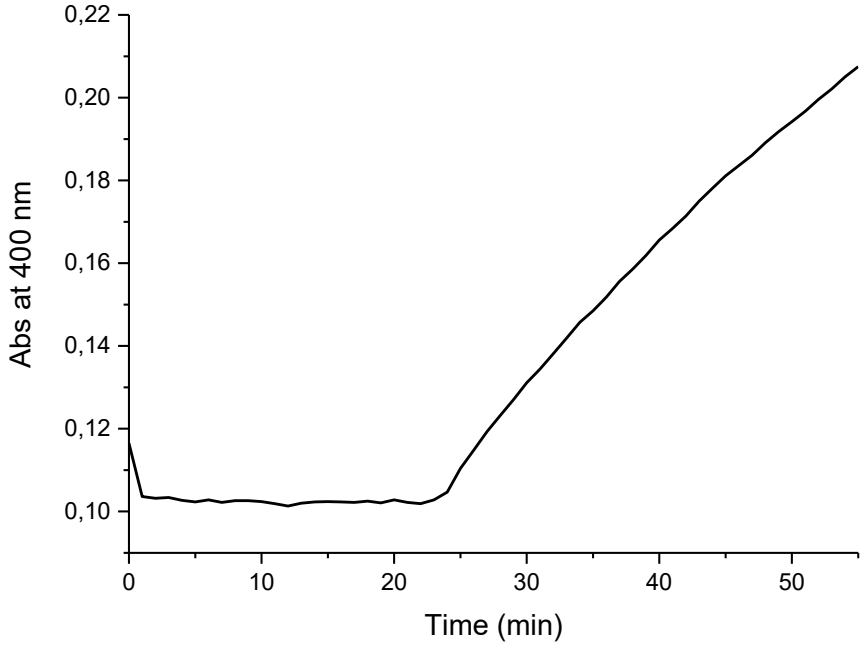


Figure S3.2 **1**-Cu^{II} 2.0 mM, HPNP 0.2 mM, TCA 15 mM, no prior pH adjustment.

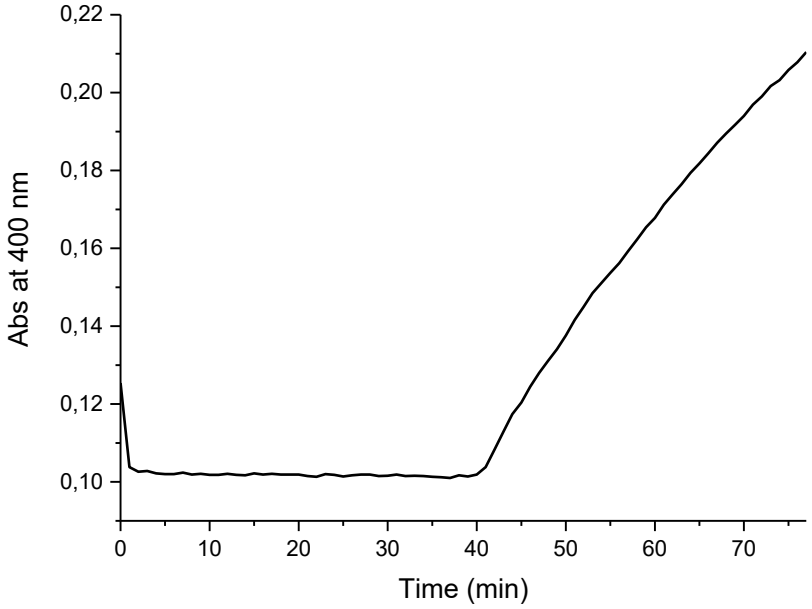


Figure S3.3 **1**-Cu^{II} 2.0 mM, HPNP 0.2 mM, TCA 20 mM, no prior pH adjustment.

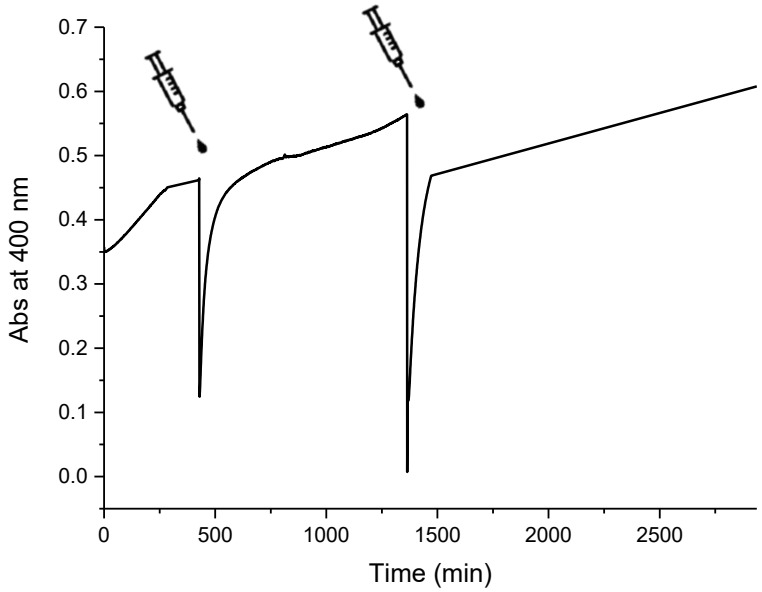


Figure S3.4 **1**-Cu^{II} 0.5 mM, HPNP 0.2 mM, TCA 10 mM; prior pH adjustment at 9.0.

This experiment, in comparison with that reported in Figure S3.5, demonstrates the influence of the catalyst concentration on the duration of the stationary state: the higher the catalyst concentration, the shorter the stationary state duration. It should be emphasized that both the occurrence and the persistence of the stationary state are highly dependent on the experimental conditions.

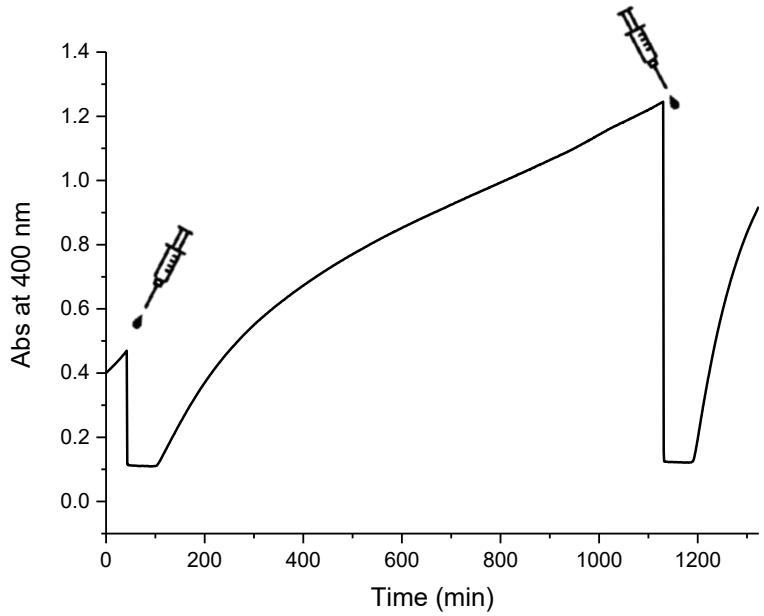


Figure S3.5 **1**-Cu^{II} 2.0 mM, HPNP 0.2 mM, TCA 10 mM; prior pH adjustment at 9.3.

This experiment, in comparison with that reported in Figure S3.4 demonstrates the influence of the catalyst concentration on the duration of the stationary state.

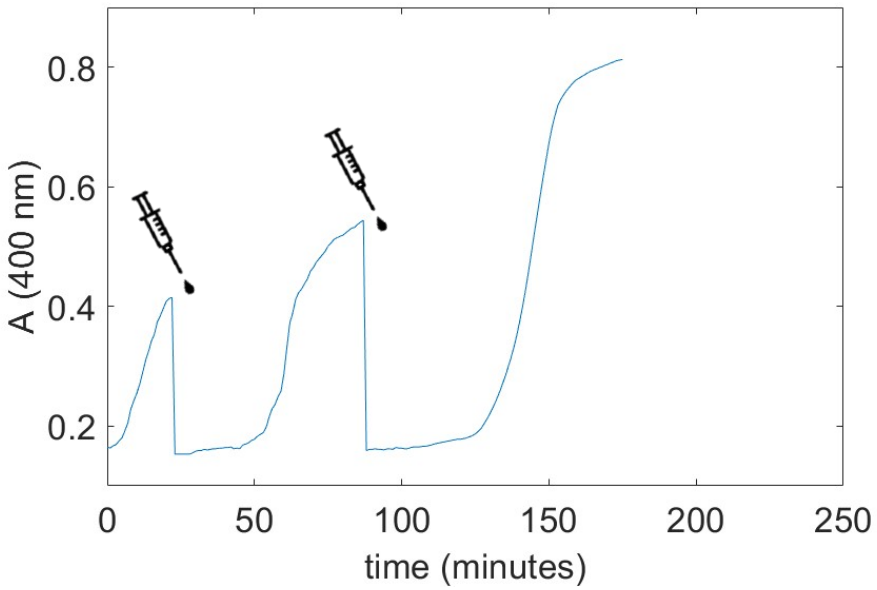


Figure S3.6 **2**-Cu^{II} 1.0 mM, HPNP 0.05 mM, TCA 10 mM, K_2CO_3 10 mM; no prior pH adjustment. The substrate was added as last component.

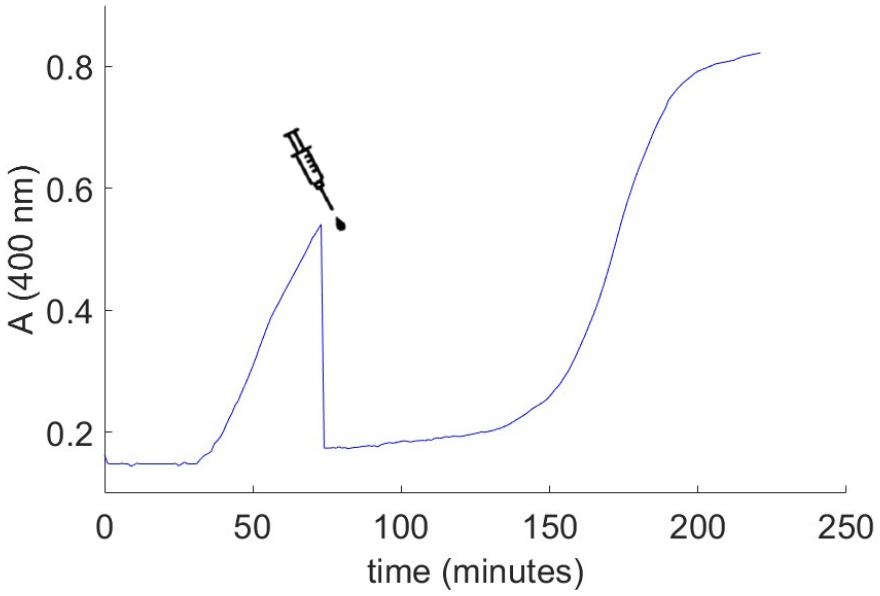


Figure S3.7 **2**-Cu^{II} 1.0 mM, HPNP 0.05 mM, TCA 20 mM, K_2CO_3 10 mM; no prior pH adjustment. The substrate was added as last component. Compare with Figure S3.6.

Section S4 pH meter calibration in 80% DMSO

The calibration of the electrode was carried out through a comparison with buffer solution of known pK_a values (see reference 58 in the main text), assumed to be the corrected pH values. The buffer solutions were mixtures of five different 10.0 mM commercial buffer acid components (see table below) and commercial 5.0 mM Tetramethylammonium hydroxide (NMe_4OH) as buffer basic component. The time elapsed to ensure a stable pH reading ranges from 5 to 10 minutes. Every measurement was repeated three times, and the obtained mean values are here reported.

Table S4.1

pH_{corr}	**pH_{read}**	**Buffer acid component**
13.83	14.55	Mesitol
11.98	12.64	*p*-chlorophenol
9.83	10.34	*p*-cianophenol
8.00	9.07	Acetic acid
3.87	4.16	Dichloroacetic acid

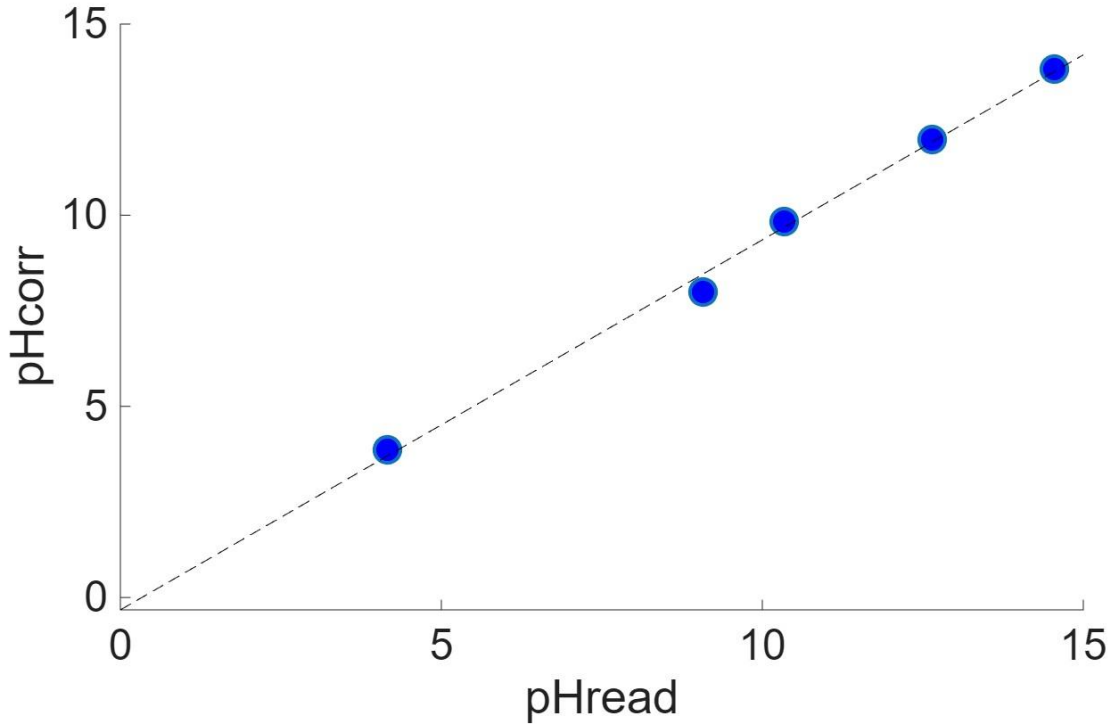


Figure S4.1 pH meter calibration curve obtained using different buffer solutions with known pK_a. See the Table reported above

Section S5 Raw Potentiometric and UV-Vis Data

TCA 10 mM, K_2CO_3 10 mM

time (min) pH_{corr}

0 12.3700000000000

1 3.29000000000000

6 3.39000000000000

11 3.48000000000000

16 3.58000000000000

21 3.71000000000000

27 3.93000000000000

32 4.28000000000000

33 4.41000000000000

34 4.55000000000000

35 4.77000000000000

36	5.18000000000000
37	6.25000000000000
38	7.61000000000000
39	7.96000000000000
41	8.25000000000000
44	8.47000000000000
51	8.76000000000000
62	8.99000000000000
66	9.04000000000000
67	3.17000000000000
76	3.22000000000000
84	3.30000000000000
97	3.48000000000000
107	3.69000000000000
113	4.13000000000000
116	4.31000000000000
118	4.44000000000000
119	4.80000000000000
121	5.16000000000000
122	5.94000000000000
123	7.24000000000000
124	7.66000000000000
125	7.85000000000000
126	8.09000000000000
128	8.47000000000000
135	8.84000000000000
152	8.87000000000000
155	3.15000000000000
156	3.21000000000000
163	3.26000000000000
167	3.41000000000000
179	3.63000000000000
190	3.92000000000000
198	4.26000000000000

203	4.48000000000000
205	5.22000000000000
208	6.02000000000000
209	7.18000000000000
210	7.54000000000000
211	7.88000000000000
213	8.06000000000000
215	8.33000000000000
220	8.66000000000000
236	8.75000000000000
243	8.85000000000000
254	8.90000000000000

1-Cu^{II} 1.0 mM, K_2CO_3 10 mM, HPNP 0.20 mM, TCA 10 mM

t(min)	A_{400nm} (a.u.)
0	0.144300000000000
1	0.138900000000000
2	0.138200000000000
3	0.138000000000000
4	0.138000000000000
5	0.138400000000000
6	0.138100000000000
7	0.136600000000000
8	0.135900000000000
9	0.135800000000000
10	0.135900000000000
11	0.136000000000000
12	0.135800000000000
13	0.136000000000000
14	0.135900000000000
15	0.136000000000000
16	0.136100000000000

17	0.136700000000000
18	0.136500000000000
19	0.136500000000000
20	0.135400000000000
21	0.134900000000000
22	0.134600000000000
23	0.134500000000000
24	0.134400000000000
25	0.134400000000000
26	0.133800000000000
27	0.134100000000000
28	0.133800000000000
29	0.134000000000000
30	0.134200000000000
31	0.134000000000000
32	0.134100000000000
33	0.134000000000000
34	0.134000000000000
35	0.134600000000000
36	0.134700000000000
37	0.134500000000000
38	0.134300000000000
39	0.134000000000000
40	0.134000000000000
41	0.133400000000000
42	0.133500000000000
43	0.133900000000000
44	0.133800000000000
45	0.133700000000000
46	0.133900000000000
47	0.133400000000000
48	0.133800000000000
49	0.133800000000000
50	0.134200000000000

51 0.133600000000000
52 0.133700000000000
53 0.134100000000000
54 0.142800000000000
55 0.140900000000000
56 0.141300000000000
57 0.141300000000000
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4279	0.265000000000000
4280	0.265600000000000
4281	0.265600000000000

1-Cu^{II} 1.0 mM, K_2CO_3 10 mM, HPNP 0.20 mM, TCA 5mM

t(min)	A_{400nm} (a.u.)
0	0.312500000000000
1	0.293000000000000
2	0.289400000000000
3	0.289800000000000
4	0.290300000000000
5	0.290500000000000
6	0.291600000000000
7	0.290800000000000
8	0.290900000000000
9	0.291600000000000
10	0.293100000000000
11	0.293500000000000
12	0.294000000000000
13	0.294800000000000
14	0.295000000000000
15	0.296100000000000
16	0.296800000000000
17	0.297100000000000

18	0.297100000000000
19	0.298100000000000
20	0.299300000000000
21	0.299400000000000
22	0.299800000000000
23	0.300100000000000
24	0.300200000000000
25	0.300800000000000
26	0.301600000000000
27	0.302200000000000
28	0.302500000000000
29	0.303400000000000
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930 0.537500000000000
931 0.537700000000000
932 0.537800000000000
933 0.538500000000000
934 0.538200000000000
935 0.539000000000000
936 0.538700000000000

937 0.538800000000000
938 0.539000000000000
939 0.539300000000000
940 0.539200000000000
941 0.539900000000000
942 0.539600000000000
943 0.539900000000000
944 0.540400000000000
945 0.540300000000000
946 0.540700000000000
947 0.540900000000000
948 0.541200000000000
949 0.541300000000000
950 0.541500000000000
951 0.541900000000000
952 0.542000000000000
953 0.542200000000000
954 0.542400000000000
955 0.542100000000000
956 0.542400000000000
957 0.543200000000000
958 0.543000000000000
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960 0.543200000000000
961 0.543400000000000
962 0.543900000000000
963 0.544100000000000
964 0.544300000000000
965 0.544600000000000
966 0.544900000000000
967 0.545000000000000
968 0.545200000000000
969 0.545400000000000
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971 0.546200000000000
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975 0.546300000000000
976 0.546500000000000
977 0.546800000000000
978 0.547000000000000
979 0.546900000000000
980 0.547100000000000
981 0.548000000000000
982 0.547900000000000
983 0.547900000000000
984 0.548500000000000
985 0.548200000000000
986 0.548700000000000
987 0.548000000000000
988 0.548300000000000
989 0.548800000000000
990 0.549000000000000
991 0.549100000000000
992 0.549300000000000
993 0.550000000000000
994 0.549800000000000
995 0.550100000000000
996 0.550200000000000
997 0.550600000000000
998 0.550700000000000
999 0.550600000000000
1000 0.550500000000000
1001 0.550900000000000
1002 0.551100000000000
1003 0.551700000000000
1004 0.551800000000000

1005 0.552000000000000
1006 0.552100000000000
1007 0.552400000000000
1008 0.552200000000000
1009 0.552400000000000
1010 0.552500000000000
1011 0.552700000000000
1012 0.553500000000000
1013 0.553500000000000
1014 0.553700000000000
1015 0.554100000000000
1016 0.553600000000000
1017 0.553900000000000
1018 0.554300000000000
1019 0.554500000000000
1020 0.554500000000000
1021 0.554600000000000
1022 0.554500000000000
1023 0.554800000000000
1024 0.555600000000000
1025 0.555800000000000
1026 0.555700000000000
1027 0.555700000000000
1028 0.555800000000000
1029 0.555900000000000
1030 0.556000000000000
1031 0.556300000000000
1032 0.556700000000000
1033 0.556900000000000
1034 0.557600000000000
1035 0.557300000000000
1036 0.557700000000000
1037 0.557600000000000
1038 0.558200000000000

1039 0.558000000000000
1040 0.558200000000000
1041 0.558400000000000
1042 0.558800000000000
1043 0.559100000000000
1044 0.558800000000000
1045 0.558800000000000
1046 0.559400000000000
1047 0.559700000000000
1048 0.560100000000000
1049 0.559800000000000
1050 0.560000000000000
1051 0.560100000000000
1052 0.560000000000000
1053 0.560400000000000
1054 0.560700000000000
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1056 0.560900000000000
1057 0.560800000000000
1058 0.561300000000000
1059 0.561700000000000
1060 0.562000000000000
1061 0.562100000000000
1062 0.562300000000000
1063 0.562300000000000
1064 0.562300000000000
1065 0.562800000000000
1066 0.562900000000000
1067 0.562700000000000
1068 0.563300000000000
1069 0.563400000000000
1070 0.563600000000000
1071 0.563700000000000
1072 0.564000000000000

1073 0.563800000000000
1074 0.564500000000000
1075 0.564300000000000
1076 0.564100000000000
1077 0.564700000000000
1078 0.564900000000000
1079 0.565000000000000
1080 0.565600000000000
1081 0.565600000000000
1082 0.565600000000000
1083 0.566100000000000
1084 0.566200000000000
1085 0.566000000000000
1086 0.566300000000000
1087 0.566100000000000
1088 0.566500000000000
1089 0.566800000000000
1090 0.566800000000000
1091 0.567400000000000
1092 0.567500000000000
1093 0.567800000000000
1094 0.567300000000000
1095 0.568500000000000
1096 0.568700000000000
1097 0.568600000000000
1098 0.568300000000000
1099 0.568300000000000
1100 0.568700000000000
1101 0.568800000000000
1102 0.569200000000000
1103 0.569500000000000
1104 0.569400000000000
1105 0.569500000000000
1106 0.570100000000000

1107 0.569800000000000
1108 0.570400000000000
1109 0.570000000000000
1110 0.570700000000000
1111 0.571000000000000
1112 0.571500000000000
1113 0.571300000000000
1114 0.571300000000000
1115 0.571300000000000
1116 0.571400000000000
1117 0.572200000000000
1118 0.572100000000000
1119 0.572500000000000
1120 0.572700000000000
1121 0.572300000000000
1122 0.572500000000000
1123 0.573000000000000
1124 0.573100000000000
1125 0.573200000000000
1126 0.573700000000000
1127 0.573700000000000
1128 0.574000000000000
1129 0.574500000000000
1130 0.574300000000000
1131 0.574600000000000
1132 0.574900000000000
1133 0.575200000000000
1134 0.575200000000000
1135 0.575300000000000
1136 0.575200000000000
1137 0.575200000000000
1138 0.576500000000000
1139 0.576100000000000
1140 0.576400000000000

1141 0.576400000000000
1142 0.576400000000000
1143 0.576600000000000
1144 0.576500000000000
1145 0.577100000000000
1146 0.577500000000000
1147 0.577200000000000
1148 0.577800000000000
1149 0.577700000000000
1150 0.578300000000000
1151 0.578800000000000
1152 0.578300000000000
1153 0.578600000000000
1154 0.578900000000000
1155 0.579000000000000
1156 0.578800000000000
1157 0.579200000000000
1158 0.579500000000000
1159 0.579700000000000
1160 0.579600000000000
1161 0.580000000000000
1162 0.580200000000000
1163 0.580500000000000
1164 0.580700000000000
1165 0.580700000000000
1166 0.581200000000000
1167 0.581000000000000
1168 0.581600000000000
1169 0.581900000000000
1170 0.581700000000000
1171 0.581800000000000
1172 0.582200000000000
1173 0.582200000000000
1174 0.582700000000000

1175 0.582700000000000
1176 0.583300000000000
1177 0.583500000000000
1178 0.583700000000000
1179 0.583700000000000
1180 0.584200000000000
1181 0.584200000000000
1182 0.584700000000000
1183 0.584700000000000
1184 0.585100000000000
1185 0.585100000000000
1186 0.585300000000000
1187 0.585500000000000
1188 0.586300000000000
1189 0.585700000000000
1190 0.586200000000000
1191 0.586900000000000
1192 0.586700000000000
1193 0.587300000000000
1194 0.587300000000000
1195 0.587300000000000
1196 0.587800000000000
1197 0.588000000000000
1198 0.588300000000000
1199 0.588600000000000
1200 0.588400000000000
1201 0.588700000000000
1202 0.588700000000000
1203 0.589300000000000
1204 0.589300000000000
1205 0.589700000000000
1206 0.590100000000000
1207 0.590800000000000
1208 0.590300000000000

1209 0.591200000000000
1210 0.591700000000000
1211 0.591200000000000
1212 0.591300000000000
1213 0.592000000000000
1214 0.591600000000000
1215 0.592300000000000
1216 0.592300000000000
1217 0.593000000000000
1218 0.592800000000000
1219 0.593600000000000
1220 0.593300000000000
1221 0.593400000000000
1222 0.594000000000000
1223 0.594000000000000
1224 0.594100000000000
1225 0.594500000000000
1226 0.594900000000000
1227 0.595200000000000
1228 0.595300000000000
1229 0.595800000000000
1230 0.596000000000000
1231 0.596000000000000
1232 0.596500000000000
1233 0.596500000000000
1234 0.596100000000000
1235 0.597100000000000
1236 0.597200000000000

1-Cu^{II} 2.0 mM, K_2CO_3 10 mM, HPNP 0.20 mM, TCA 10 mM, no pH adjustment.
See Fig. 5 in the main text.

t(min) A_{400nm} (a.u.)

0	0.126400000000000
1	0.116900000000000
2	0.121600000000000
3	0.125000000000000
4	0.129600000000000
5	0.133200000000000
6	0.137100000000000
7	0.141000000000000
8	0.144600000000000
9	0.148100000000000
10	0.151800000000000
11	0.155200000000000
12	0.158400000000000
13	0.161900000000000
14	0.165300000000000
15	0.169200000000000
16	0.172200000000000
17	0.176100000000000
18	0.180100000000000
19	0.182700000000000
20	0.186400000000000
21	0.189900000000000
22	0.193000000000000
23	0.196100000000000
24	0.199900000000000
25	0.203200000000000
26	0.206700000000000
27	0.209600000000000
28	0.212600000000000
29	0.215800000000000
30	0.218800000000000
31	0.221900000000000

1-Cu^{II} 2.0 mM, K_2CO_3 10 mM, HPNP 0.20 mM, TCA 15 mM, no pH adjustment.
See Fig. 5 in the main text.

t(min)	A_{400nm} (a.u.)
0	0.116500000000000
1	0.103600000000000
2	0.103200000000000
3	0.103400000000000
4	0.102700000000000
5	0.102300000000000
6	0.102800000000000
7	0.102200000000000
8	0.102600000000000
9	0.102600000000000
10	0.102400000000000
11	0.101900000000000
12	0.101300000000000
13	0.102000000000000
14	0.102300000000000
15	0.102400000000000
16	0.102300000000000
17	0.102200000000000
18	0.102500000000000
19	0.102100000000000
20	0.102800000000000
21	0.102200000000000
22	0.101900000000000
23	0.102800000000000
24	0.104700000000000
25	0.110400000000000
26	0.114800000000000
27	0.119300000000000
28	0.123200000000000
29	0.127000000000000

30	0.131100000000000
31	0.134400000000000
32	0.138100000000000
33	0.141900000000000
34	0.145700000000000
35	0.148500000000000
36	0.151800000000000
37	0.155600000000000
38	0.158600000000000
39	0.161900000000000
40	0.165600000000000
41	0.168400000000000
42	0.171400000000000
43	0.175000000000000
44	0.178100000000000
45	0.181100000000000
46	0.183600000000000
47	0.186100000000000
48	0.189100000000000
49	0.191800000000000
50	0.194200000000000
51	0.196700000000000
52	0.199600000000000
53	0.202100000000000
54	0.205000000000000
55	0.207500000000000

1-Cu^{II} 2.0 mM, K_2CO_3 10 mM, HPNP 0.20 mM, TCA 20 mM, no pH adjustment.
See Fig. 5 in the main text.

t(min)	A_{400nm} (a.u.)
0	0.125400000000000

1	0.103800000000000
2	0.102600000000000
3	0.102800000000000
4	0.102200000000000
5	0.102000000000000
6	0.102000000000000
7	0.102400000000000
8	0.101900000000000
9	0.102100000000000
10	0.101800000000000
11	0.101800000000000
12	0.102100000000000
13	0.101800000000000
14	0.101700000000000
15	0.102200000000000
16	0.101900000000000
17	0.102100000000000
18	0.101900000000000
19	0.101900000000000
20	0.101900000000000
21	0.101500000000000
22	0.101300000000000
23	0.102000000000000
24	0.101800000000000
25	0.101400000000000
26	0.101700000000000
27	0.101900000000000
28	0.101900000000000
29	0.101500000000000
30	0.101600000000000
31	0.101900000000000
32	0.101500000000000
33	0.101600000000000
34	0.101500000000000

35 0.101300000000000
36 0.101200000000000
37 0.101000000000000
38 0.101700000000000
39 0.101400000000000
40 0.101900000000000
41 0.103800000000000
42 0.108300000000000
43 0.112900000000000
44 0.117400000000000
45 0.120400000000000
46 0.124500000000000
47 0.128000000000000
48 0.131100000000000
49 0.134100000000000
50 0.137600000000000
51 0.141600000000000
52 0.145000000000000
53 0.148500000000000
54 0.151100000000000
55 0.153700000000000
56 0.156200000000000
57 0.159300000000000
58 0.162300000000000
59 0.165400000000000
60 0.167800000000000
61 0.171200000000000
62 0.173900000000000
63 0.176500000000000
64 0.179400000000000
65 0.181800000000000
66 0.184400000000000
67 0.187100000000000
68 0.189500000000000

69	0.191700000000000
70	0.194000000000000
71	0.196900000000000

2-Cu^{II} xxxxxx mM, K_2CO_3 10 mM, HPNP 50 μM, TCA 10 mM, no prior pH adjustment.
See Fig. 6 in the main text.

t(min)	A_{400nm} (a.u.)
0	0.165000000000000
1	0.163000000000000
2	0.167000000000000
3	0.169000000000000
4	0.175000000000000
5	0.180000000000000
6	0.192000000000000
7	0.205000000000000
8	0.228000000000000
9	0.242000000000000
10	0.254000000000000
11	0.269000000000000
12	0.289000000000000
13	0.309000000000000
14	0.324000000000000
15	0.342000000000000
16	0.353000000000000
17	0.374000000000000
18	0.384000000000000
19	0.396000000000000
20	0.408000000000000
21	0.413000000000000
22	0.415000000000000
23	0.153000000000000
24	0.153000000000000

25 0.153000000000000
26 0.153000000000000
27 0.153000000000000
28 0.153000000000000
29 0.155000000000000
30 0.158000000000000
31 0.159000000000000
32 0.159000000000000
33 0.161000000000000
34 0.160000000000000
35 0.161000000000000
36 0.162000000000000
37 0.162000000000000
38 0.163000000000000
39 0.163000000000000
40 0.164000000000000
41 0.164000000000000
42 0.165000000000000
43 0.162000000000000
44 0.163000000000000
45 0.162000000000000
46 0.168000000000000
47 0.170000000000000
48 0.172000000000000
49 0.176000000000000
50 0.178000000000000
51 0.183000000000000
52 0.186000000000000
53 0.189000000000000
54 0.198000000000000
55 0.215000000000000
56 0.229000000000000
57 0.236000000000000
58 0.249000000000000

59 0.258000000000000
60 0.287000000000000
61 0.331000000000000
62 0.372000000000000
63 0.389000000000000
64 0.412000000000000
65 0.423000000000000
66 0.429000000000000
67 0.438000000000000
68 0.446000000000000
69 0.459000000000000
70 0.465000000000000
71 0.474000000000000
72 0.480000000000000
73 0.486000000000000
74 0.492000000000000
75 0.501000000000000
76 0.506000000000000
77 0.512000000000000
78 0.515000000000000
79 0.517000000000000
80 0.519000000000000
81 0.523000000000000
82 0.527000000000000
83 0.531000000000000
84 0.532000000000000
85 0.537000000000000
86 0.541000000000000
87 0.544000000000000
88 0.159000000000000
89 0.161000000000000
90 0.161000000000000
91 0.162000000000000
92 0.161000000000000

93 0.161000000000000
94 0.160000000000000
95 0.162000000000000
96 0.162000000000000
97 0.161000000000000
98 0.164000000000000
99 0.163000000000000
100 0.163000000000000
101 0.162000000000000
102 0.162000000000000
103 0.164000000000000
104 0.165000000000000
105 0.165000000000000
106 0.165000000000000
107 0.166000000000000
108 0.166000000000000
109 0.167000000000000
110 0.168000000000000
111 0.170000000000000
112 0.171000000000000
113 0.172000000000000
114 0.173000000000000
115 0.174000000000000
116 0.175000000000000
117 0.176000000000000
118 0.177000000000000
119 0.178000000000000
120 0.178000000000000
121 0.179000000000000
122 0.180000000000000
123 0.182000000000000
124 0.184000000000000
125 0.187000000000000
126 0.191000000000000

127 0.196000000000000
128 0.204000000000000
129 0.212000000000000
130 0.221000000000000
131 0.231000000000000
132 0.242000000000000
133 0.254000000000000
134 0.267000000000000
135 0.281000000000000
136 0.296000000000000
137 0.312000000000000
138 0.330000000000000
139 0.350000000000000
140 0.373000000000000
141 0.398000000000000
142 0.426000000000000
143 0.456000000000000
144 0.488000000000000
145 0.521000000000000
146 0.552000000000000
147 0.584000000000000
148 0.615000000000000
149 0.644000000000000
150 0.673000000000000
151 0.698000000000000
152 0.718000000000000
153 0.736000000000000
154 0.746000000000000
155 0.754000000000000
156 0.761000000000000
157 0.767000000000000
158 0.773000000000000
159 0.778000000000000
160 0.781000000000000

161	0.784000000000000
162	0.787000000000000
163	0.790000000000000
164	0.793000000000000
165	0.795000000000000
166	0.797000000000000
167	0.799000000000000
168	0.801000000000000
169	0.803000000000000
170	0.805000000000000
171	0.807000000000000
172	0.809000000000000
173	0.811000000000000
174	0.812000000000000
175	0.813000000000000

2-Cu^{II} xxxxx mM, K_2CO_3 10 mM, HPNP 50 μM, TCA 20 mM, no prior pH adjustment.
See Fig. 6 in the main text.

t(min)	A_{400nm} (a.u.)
0	0.163000000000000
1	0.149000000000000
2	0.148000000000000
3	0.148000000000000
4	0.148000000000000
5	0.148000000000000
6	0.149000000000000
7	0.148000000000000
8	0.148000000000000
9	0.144000000000000
10	0.147000000000000
11	0.149000000000000

12	0.148000000000000
13	0.148000000000000
14	0.148000000000000
15	0.148000000000000
16	0.148000000000000
17	0.148000000000000
18	0.148000000000000
19	0.148000000000000
20	0.148000000000000
21	0.148000000000000
22	0.148000000000000
23	0.148000000000000
24	0.149000000000000
25	0.145000000000000
26	0.148000000000000
27	0.151000000000000
28	0.148000000000000
29	0.148000000000000
30	0.148000000000000
31	0.148000000000000
32	0.154000000000000
33	0.159000000000000
34	0.163000000000000
35	0.165000000000000
36	0.169000000000000
37	0.181000000000000
38	0.186000000000000
39	0.192000000000000
40	0.201000000000000
41	0.212000000000000
42	0.223000000000000
43	0.232000000000000
44	0.244000000000000
45	0.251000000000000

46 0.263000000000000
47 0.274000000000000
48 0.285000000000000
49 0.297000000000000
50 0.309000000000000
51 0.322000000000000
52 0.336000000000000
53 0.349000000000000
54 0.362000000000000
55 0.375000000000000
56 0.387000000000000
57 0.397000000000000
58 0.406000000000000
59 0.415000000000000
60 0.425000000000000
61 0.434000000000000
62 0.443000000000000
63 0.452000000000000
64 0.461000000000000
65 0.470000000000000
66 0.479000000000000
67 0.488000000000000
68 0.497000000000000
69 0.507000000000000
70 0.518000000000000
71 0.525000000000000
72 0.533000000000000
73 0.541000000000000
74 0.174000000000000
75 0.174000000000000
76 0.174000000000000
77 0.175000000000000
78 0.174000000000000
79 0.176000000000000

80	0.174000000000000
81	0.175000000000000
82	0.173000000000000
83	0.174000000000000
84	0.175000000000000
85	0.175000000000000
86	0.176000000000000
87	0.177000000000000
88	0.177000000000000
89	0.178000000000000
90	0.177000000000000
91	0.178000000000000
92	0.176000000000000
93	0.179000000000000
94	0.181000000000000
95	0.182000000000000
96	0.183000000000000
97	0.182000000000000
98	0.183000000000000
99	0.184000000000000
100	0.185000000000000
101	0.186000000000000
102	0.185000000000000
103	0.184000000000000
104	0.185000000000000
105	0.186000000000000
106	0.186000000000000
107	0.187000000000000
108	0.186000000000000
109	0.188000000000000
110	0.187000000000000
111	0.189000000000000
112	0.191000000000000
113	0.190000000000000

114 0.191000000000000
115 0.193000000000000
116 0.192000000000000
117 0.193000000000000
118 0.194000000000000
119 0.193000000000000
120 0.193000000000000
121 0.194000000000000
122 0.195000000000000
123 0.196000000000000
124 0.197000000000000
125 0.198000000000000
126 0.199000000000000
127 0.201000000000000
128 0.200000000000000
129 0.201000000000000
130 0.202000000000000
131 0.203000000000000
132 0.205000000000000
133 0.206000000000000
134 0.208000000000000
135 0.210000000000000
136 0.212000000000000
137 0.215000000000000
138 0.218000000000000
139 0.221000000000000
140 0.224000000000000
141 0.227000000000000
142 0.230000000000000
143 0.234000000000000
144 0.238000000000000
145 0.241000000000000
146 0.244000000000000
147 0.247000000000000

148 0.250000000000000
149 0.254000000000000
150 0.259000000000000
151 0.265000000000000
152 0.271000000000000
153 0.276000000000000
154 0.283000000000000
155 0.291000000000000
156 0.298000000000000
157 0.307000000000000
158 0.317000000000000
159 0.327000000000000
160 0.338000000000000
161 0.349000000000000
162 0.360000000000000
163 0.372000000000000
164 0.384000000000000
165 0.396000000000000
166 0.409000000000000
167 0.423000000000000
168 0.438000000000000
169 0.454000000000000
170 0.471000000000000
171 0.488000000000000
172 0.506000000000000
173 0.524000000000000
174 0.542000000000000
175 0.559000000000000
176 0.575000000000000
177 0.590000000000000
178 0.605000000000000
179 0.619000000000000
180 0.632000000000000
181 0.644000000000000

182 0.656000000000000
183 0.668000000000000
184 0.680000000000000
185 0.692000000000000
186 0.703000000000000
187 0.713000000000000
188 0.722000000000000
189 0.731000000000000
190 0.744000000000000
191 0.751000000000000
192 0.757000000000000
193 0.763000000000000
194 0.768000000000000
195 0.773000000000000
196 0.777000000000000
197 0.781000000000000
198 0.785000000000000
199 0.789000000000000
200 0.792000000000000
201 0.794000000000000
202 0.796000000000000
203 0.798000000000000
204 0.800000000000000
205 0.802000000000000
206 0.804000000000000
207 0.805000000000000
208 0.806000000000000
209 0.807000000000000
210 0.808000000000000
211 0.809000000000000
212 0.810000000000000
213 0.812000000000000
214 0.814000000000000
215 0.816000000000000

216 0.817000000000000

217 0.818000000000000

218 0.819000000000000

219 0.820000000000000

220 0.821000000000000

221 0.822000000000000

S6 Potentiometric data from previous investigations

The following data are taken from our previous investigation *J. Org. Chem* 2015, 80, 5887 and *J. Org. Chem* 2011, 76, 5438 (cited as references 43 and 65 in the main text). A reviewer asked details about the species present in solution. Therefore, we report these data for the reader convenience.

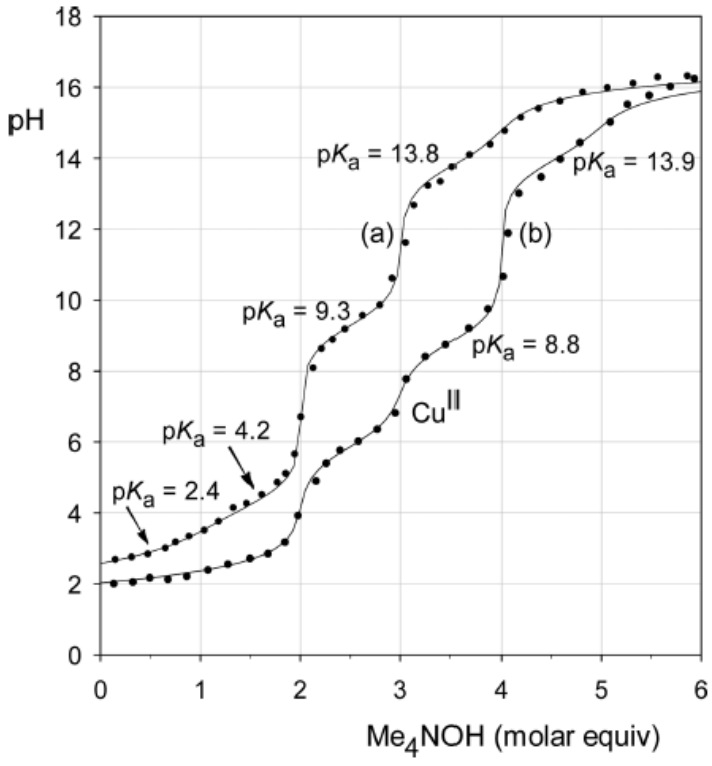


Figure S6.1 Potentiometric titration of 1.0 mM **2** with Me_4NOH in 80% DMSO in the absence (a) and presence (b) of 1 molar equiv of Cu^{II}. Data points are experimental and the lines are calculated. The Log $K_{Cu} = 9.2$

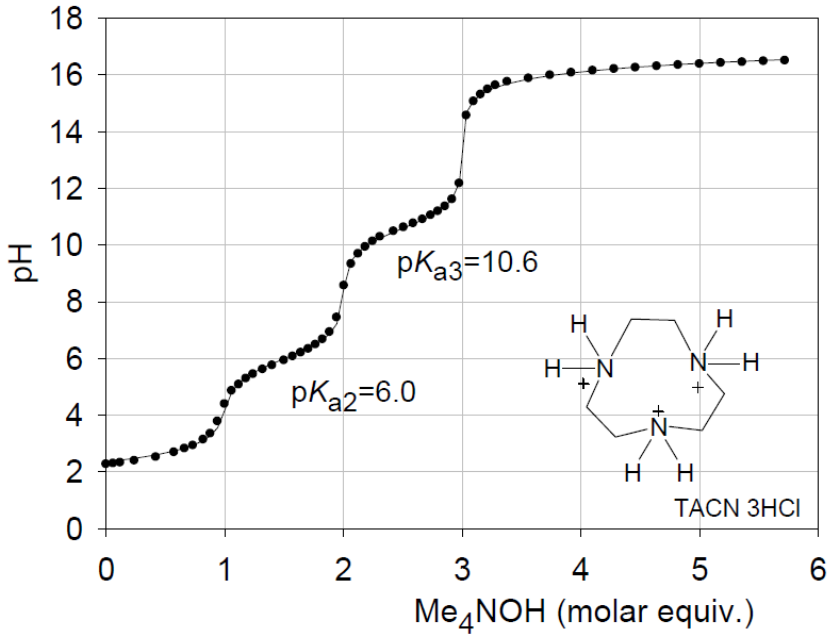


Figure S6.2 Potentiometric titration of 1.0 mM TACN with Me_4NOH in 80% DMSO

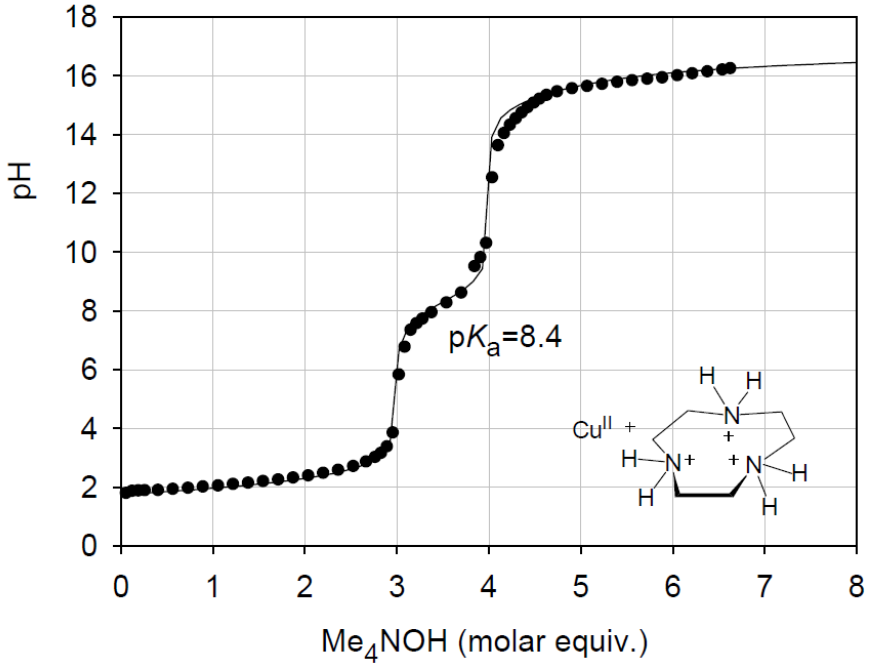


Figure S6.3 Potentiometric titration of 1.0 mM TACN with Me_4NOH in 80% DMSO in the presence of equimolar amount of Cu^{II}.Log K_{Cu} = 11.3