

Supplementary Information

Evaluation and optimization of pilot-scale catalytic ozonation - persulfate oxidation integrated process for the pretreatment of dry-spun acrylic fiber wastewater

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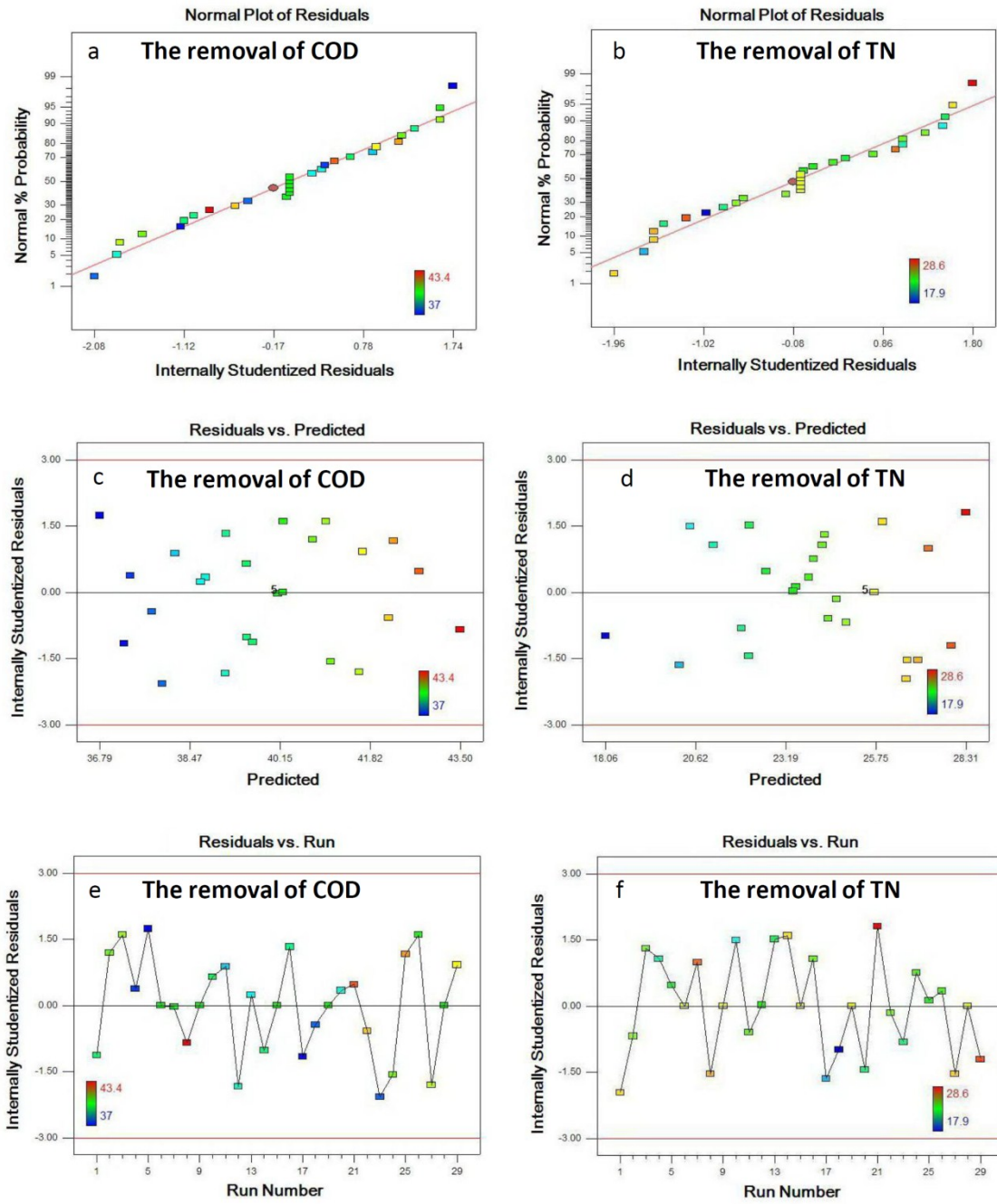


Figure S1 Residual plots for COD (a, b, c) and TN (d, e, f) removal efficiency of DAF wastewater.

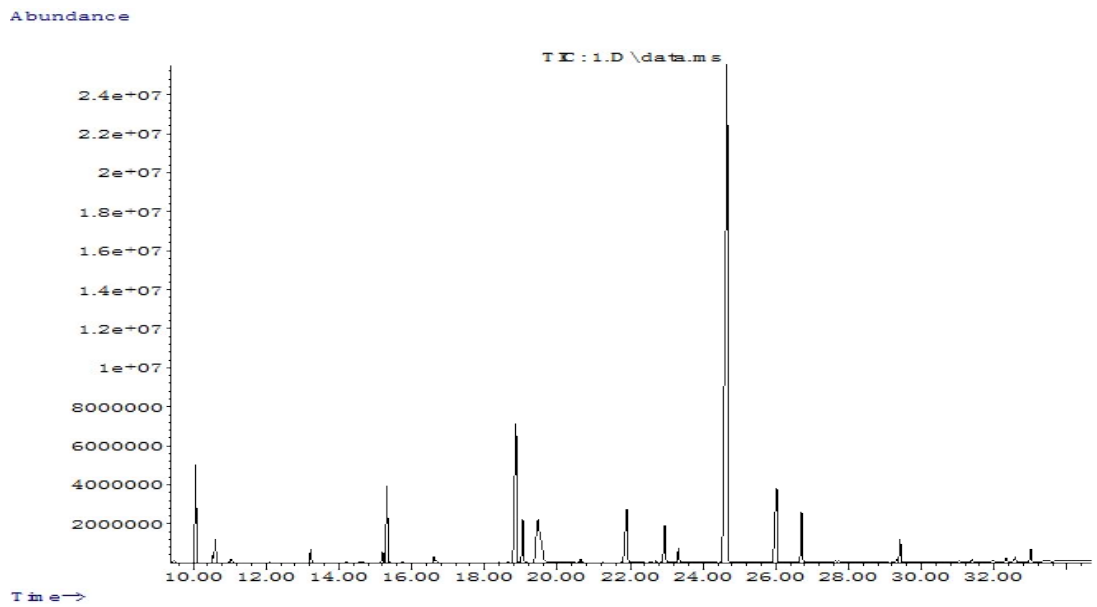


Fig S2 Total ion current chromatogram of the initial DAF wastewater.

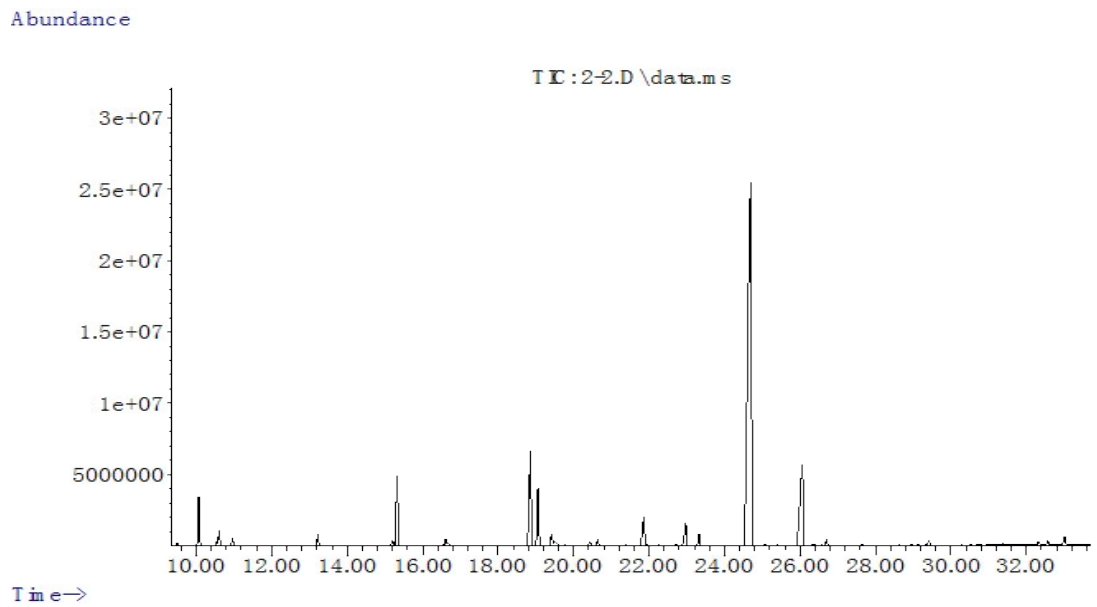


Fig S3 Total ion current chromatogram of wastewater after catalytic oxidation treatment.

Table S1 The design of RSM and its actual and predicted values.

Number	Reaction time (h)	Reaction temperature (°C)	The addition of ozone (g/h)	The addition of persulfate (kg/t)	The removal of COD (%)		The removal of TN (%)	
					Experimental	Predicted	Experimental	Predicted
1	5.00	70.00	30.00	1.30	39.5	39.64	26.3	26.61
2	6.00	60.00	30.00	0.50	40.9	40.75	24.8	24.91
3	5.00	70.00	40.00	0.90	41.2	41.0	24.5	24.29
4	4.00	60.00	20.00	0.90	37.4	37.35	21.3	21.13
5	5.00	70.00	20.00	0.90	37.0	36.79	22.7	22.63
6	5.00	60.00	30.00	0.90	40.2	40.2	25.7	25.7
7	4.00	60.00	30.00	1.30	40.1	40.10	27.4	27.24
8	6.00	60.00	40.00	0.90	43.4	43.5	26.7	26.95
9	5.00	60.00	30.00	0.90	40.2	40.2	25.7	25.7
10	5.00	50.00	30.00	0.50	39.6	39.52	20.7	20.46
11	4.00	70.00	30.00	0.90	38.3	38.19	24.3	24.4
12	5.00	60.00	20.00	1.30	38.9	39.13	23.4	23.4
13	4.00	60.00	30.00	0.50	38.7	38.67	22.4	22.16
14	6.00	70.00	30.00	0.90	39.4	39.52	26.2	25.95
15	5.00	60.00	30.00	0.90	40.2	40.2	25.7	25.7
16	6.00	60.00	20.00	0.90	39.3	39.14	24.4	24.23
17	5.00	60.00	20.00	0.50	37.1	37.24	19.9	20.16
18	5.00	50.00	20.00	0.90	37.7	37.75	17.9	18.06
19	5.00	60.00	30.00	0.90	40.2	40.2	25.7	25.7
20	4.00	50.00	30.00	0.90	38.8	38.76	21.9	22.13
21	5.00	60.00	40.00	1.30	42.8	42.74	28.6	28.31
22	5.00	50.00	40.00	0.90	42.1	42.17	24.6	24.63
23	5.00	70.00	30.00	0.50	37.7	37.95	21.8	21.93
24	6.00	50.00	30.00	0.90	40.9	41.09	24.1	23.98
25	5.00	60.00	40.00	0.50	42.4	42.26	23.5	23.48
26	5.00	50.00	30.00	1.30	40.4	40.2	23.9	23.85
27	4.00	60.00	40.00	0.90	41.4	41.62	26.4	26.65
28	5.00	60.00	30.00	0.90	40.2	40.2	25.7	25.7
29	6.00	60.00	30.00	1.30	41.8	41.69	27.7	27.89

Table S2 Analysis of variance table for the removal of COD.

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob> F
Model	80.77	14	5.77	160.74	< 0.0001
A-Reaction time	10.08	1	10.08	280.93	< 0.0001
B-Reaction temperature	3.41	1	3.41	95.10	< 0.0001
C-The addition of ozone	55.90	1	55.90	1557.44	< 0.0001
D-The addition of persulfate	4.20	1	4.20	117.04	< 0.0001
AB	0.25	1	0.25	6.97	0.0194
AC	2.500E-003	1	2.500E-003	0.070	0.7957
AD	0.063	1	0.063	1.74	0.2081
BC	0.010	1	0.010	0.28	0.6059
BD	0.25	1	0.25	6.97	0.0194
CD	0.49	1	0.49	13.65	0.0024
A ²	0.045	1	0.045	1.25	0.2815
B ²	5.16	1	5.16	143.68	< 0.0001
C ²	0.095	1	0.095	2.64	0.1266
D ²	2.815E-003	1	2.815E-003	0.078	0.7835
Residual	0.50	14	0.036		
Lack of Fit	0.50	10	0.050		
Pure Error	0.000	4	0.000		
Cor Total	81.27	28			
Std. Dev.	0.19	R-Squared	0.9938		
Mean	19.92	Adj R-Squared	0.9876		
C.V. %	0.95	Pred R-Squared	0.9644		
PRESS	2.89	Adeq Precision	49.295		

Table S3 Analysis of variance table for the removal of TN.

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob> F
Model	168.87	14	12.06	198.08	< 0.0001
A-Reaction time	8.67	1	8.67	142.38	< 0.0001
B-Reaction temperature	13.44	1	13.44	220.73	< 0.0001
C-The addition of ozone	50.84	1	50.84	834.92	< 0.0001
D-The addition of persulfate	48.80	1	48.80	801.46	< 0.0001
AB	0.023	1	0.023	0.37	0.5530
AC	1.96	1	1.96	32.19	< 0.0001
AD	1.10	1	1.10	18.11	0.0008
BC	6.00	1	6.00	98.57	< 0.0001
BD	0.42	1	0.42	6.94	0.0196
CD	0.64	1	0.64	10.51	0.0059
A ²	0.91	1	0.91	14.98	0.0017
B ²	24.98	1	24.98	410.26	< 0.0001
C ²	11.60	1	11.60	190.56	< 0.0001
D ²	1.79	1	1.79	29.36	< 0.0001
Residual	0.85	14	0.061		
Lack of Fit	0.85	10	0.085		
Pure Error	0.000	4	0.000		
Cor Total	169.72	28			
Std. Dev.	0.25	R-Squared	0.9950		
Mean	24.27	Adj R-Squared	0.9900		
C.V. %	1.02	Pred R-Squared	0.9711		
PRESS	4.91	Adeq Precision	57.779		

Table S4 Chemical composition analysis of the initial DAF wastewater.

No	Compound	Retention Time (min)	Formula	Peak area	March
1	Ethyl Acetate	10.06	C ₄ H ₈ O ₂	139914155	95.4
2	Trichloromethane	10.53	CHCl ₃	11780263	97.7
3	Tetrahydrofuran	10.60	C ₄ H ₈ O	34347508	95.9
4	2,2-Dimethoxy-propane	10.96	C ₅ H ₁₂ O ₂	1279925	88
5	Acetic acid	11.05	C ₂ H ₄ O ₂	6591368	95.2
6	1,1-Dimethoxy-propane	12.09	C ₅ H ₁₂ O ₂	760356	84
7	1-Butanol	12.30	C ₄ H ₁₀ O	298596	87.2
8	Dimethoxy-methane	13.22	C ₃ H ₈ O ₂	18385137	85.7
9	Propanoic acid	14.18	C ₃ H ₆ O ₂	1213664	86.5
10	2-Methoxytetrahydrofuran	14.54	C ₅ H ₁₀ O ₂	757438	89.2
11	2-Propenoic acid	14.61	C ₃ H ₄ O ₂	3342516	92.8
12	Toluene	15.19	C ₇ H ₈	17253537	95.4
13	1,3-Diazine	15.32	C ₄ H ₄ N ₂	118582547	96
14	Tetraethylene glycol	15.75	C ₈ H ₁₈ O ₅	793473	77
15	Acetic acid, butyl ester	16.61	C ₆ H ₁₂ O ₂	8582803	96.5
16	4-Methyl-3-penten-2-one	16.67	C ₆ H ₁₀ O	3125538	91.5
17	Ethylbenzene	18.40	C ₈ H ₁₀	537918	85.8
18	1,3-Dimethyl-benzene	18.65	C ₈ H ₁₀	1151045	92.4
19	(E)-2-Butenedinitrile	18.86	C ₄ H ₂ N ₂	291358469	96.3
20	4-Hydroxy-4-methyl-2-pentanone	19.05	C ₆ H ₁₂ O ₂	68007131	92.7
21	1H-Pyrazole	19.47	C ₃ H ₄ N ₂	204894871	97.2
22	4-Methoxy-4-methyl-2-pentanone	20.64	C ₇ H ₁₄ O ₂	7568376	95
23	4-Oxobutanenitrile	21.90	C ₄ H ₅ NO	128224798	91.6
24	2-Ethyl-2,4,5-trimethyl-1,3-dioxolane	22.27	C ₈ H ₁₆ O ₂	1404986	64.8
25	3-Aminoacrylonitriles	22.71	C ₃ H ₄ N ₂	2917493	83.7
26	Methylene-propanedinitrile	22.95	C ₄ H ₂ N ₂	66651689	90.2
27	4-Pyridinecarbonitrile	23.32	C ₆ H ₄ N ₂	21560069	96.6
28	Acetyl	23.75	C ₁₁ H ₁₂ N ₂	966745	80.3
29	3-Pyridinecarbonitrile	24.66	C ₆ H ₄ N ₂	1272910771	95.6
30	2-Pentenedinitrile	24.86	C ₅ H ₄ N ₂	884316	79.4
31	2-Butanone, 3-hydroxy-	25.07	C ₄ H ₈ O ₂	1338502	90.1
32	Pentanedinitrile	25.39	C ₅ H ₆ N ₂	3419030	92.3
33	Butanedinitrile	26.02	C ₄ H ₄ N ₂	329529167	96.3
34	2,3-Dimethyl- butanedinitrile	26.38	C ₆ H ₈ N ₂	4389537	86.6
35	2-Methyl- pentanedinitrile	26.57	C ₆ H ₈ N ₂	2405294	72.9
36	2-Pyrimidinol	26.71	C ₄ H ₄ N ₂ O	75919917	81.3
37	4-(Aminomethyl)pyridine	27.64	C ₆ H ₈ N ₂	1753546	71.8

38	4-Aminonicotinonitrile	27.73	C ₆ H ₅ N ₃	1695558	67.3
39	5-Azabenzimidazole	29.32	C ₆ H ₅ N ₃	3310281	85.2
40	3-Imidazol-1-ylpropanenitrile	29.42	C ₆ H ₇ N ₃	35067015	87.9
41	3-Pyridineacetonitrile	31.39	C ₇ H ₆ N ₂	4098948	93.7
42	3-Hexenedinitrile	31.56	C ₆ H ₆ N ₂	1183489	91.8
43	1,3,8-Triazanaphthalene	31.96	C ₇ H ₅ N ₃	2979977	81.9
44	1,4-Benzenedicarbonitrile	32.31	C ₈ H ₄ N ₂	8385614	94.5
45	1,3-Benzenedicarbonitrile	32.57	C ₈ H ₄ N ₂	8464372	96.1
46	2,3-Dicyanopropionamide	33.00	C ₅ H ₅ N ₃ O	18948088	85.1

Table S5 Chemical composition analysis of wastewater after catalytic oxidation treatment.

No	Compound	Retention Time (min)	Formula	Peak area	March
1	Ethyl Acetate	10.07	C ₄ H ₈ O ₂	93409141	95.4
2	Trichloromethane	10.53	CHCl ₃	7035379	97.7
3	Tetrahydrofuran	10.61	C ₄ H ₈ O	29151671	95.9
4	2,2-Dimethoxy-propane	10.96	C ₅ H ₁₂ O ₂	14669281	88
5	2-(1-methylethoxy)- Ethanol	11.08	C ₅ H ₁₂ O ₂	640176	70.8
6	Acetic acid	12.09	C ₂ H ₄ O ₂	452463	95.2
7	1,1-Dimethoxy-propane	12.30	C ₅ H ₁₂ O ₂	429236	84
8	1-Butanol	13.22	C ₄ H ₁₀ O	22400193	87.2
9	Dimethoxy-methane	13.93	C ₃ H ₈ O ₂	594527	85.7
10	2-Methoxytetrahydrofuran	14.54	C ₅ H ₁₀ O ₂	840538	89.2
11	Toluene	15.19	C ₇ H ₈	11260853	95.4
12	1,3-Diazine	15.32	C ₄ H ₄ N ₂	156252487	96
13	Tetraethylene glycol	15.75	C ₈ H ₁₈ O ₅	839353	77
14	Formamide	15.88	CH ₃ NO	1270827	96.2
15	3-Nitropropanoic acid	16.55	C ₃ H ₅ NO ₄	2111076	82.3
16	Acetic acid, butyl ester	16.61	C ₆ H ₁₂ O ₂	12433965	96.5
17	4-Methyl-3-penten-2-one	16.67	C ₆ H ₁₀ O	4894581	91.5
18	2-Ethoxy-2-methoxy- propane	17.68	C ₆ H ₁₄ O ₂	713202	72.1
19	Ethylbenzene	18.40	C ₈ H ₁₀	697061	85.8
20	1,3-Dimethyl-benzene	18.65	C ₈ H ₁₀	1390882	92.4
21	(E)-2-Butenedinitrile	18.86	C ₄ H ₂ N ₂	195248668	96.3
22	4-Hydroxy-4-methyl-2-pentanone	19.06	C ₆ H ₁₂ O ₂	129529727	92.7
23	1H-Pyrazole	19.41	C ₃ H ₄ N ₂	44555379	97.2
24	2-Hydroxy-propanenitrile	19.75	C ₃ H ₅ NO	2814513	89.6
25	Hydroxy-acetonitrile	20.43	C ₂ H ₃ NO	10486104	91.8
26	4-Methoxy-4-methyl-2-pentanone	20.64	C ₇ H ₁₄ O ₂	13484907	95
27	3-Hydroxy- propanenitrile	21.38	C ₃ H ₅ NO	2838458	92.6
28	4-Oxobutanenitrile	21.86	C ₄ H ₅ NO	86391572	91.6
29	2-Ethyl-2,4,5-trimethyl-1,3-dioxolane	22.27	C ₈ H ₁₆ O ₂	1317870	64.8
30	3-Aminoacrylonitriles	22.72	C ₃ H ₄ N ₂	1673115	83.7
31	Methylene-propanedinitrile	22.97	C ₄ H ₂ N ₂	50292200	90.2
32	4-Pyridinecarbonitrile	23.32	C ₆ H ₄ N ₂	18762264	96.6
33	Acetyl	23.75	C ₁₁ H ₁₂ N ₂	636939	80.3
34	3-Pyridinecarbonitrile	24.69	C ₆ H ₄ N ₂	920290854	95.6
35	2-Pentenedinitrile	24.87	C ₅ H ₄ N ₂	1303435	79.4
36	2-Butanone, 3-hydroxy-	25.07	C ₄ H ₈ O ₂	3650907	90.1
37	Pentanedinitrile	25.40	C ₅ H ₆ N ₂	1527576	92.3

38	Butanedinitrile	26.06	C ₄ H ₄ N ₂	173724896	96.3
39	2,3-Dimethyl- butanedinitrile	26.39	C ₆ H ₈ N ₂	1140163	86.6
40	2-Methyl- pentanedinitrile	26.57	C ₆ H ₈ N ₂	750944	72.9
41	2-Pyrimidinol	26.70	C ₄ H ₄ N ₂ O	13599489	81.3
42	4-(Aminomethyl)pyridine	27.64	C ₆ H ₈ N ₂	2807082	71.8
43	3-Imidazol-1-ylpropanenitrile	29.41	C ₆ H ₇ N ₃	8745363	87.9
44	3-Pyridineacetonitrile	31.39	C ₇ H ₆ N ₂	3206225	93.7
45	1,3,8-Triazanaphthalene	31.96	C ₇ H ₅ N ₃	1384103	81.9
46	1,4-Benzenedicarbonitrile	32.32	C ₈ H ₄ N ₂	6759239	94.5
47	1,3-Benzenedicarbonitrile	32.57	C ₈ H ₄ N ₂	6676888	96.1
48	2,3-Dicyanopropionamide	33.00	C ₅ H ₅ N ₃ O	18812023	85.1
