

## Sustainable hydrothermal carbon for advanced electrochemical energy storage

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**Supplementary**



**Table S1** Physicochemical properties of raw hydrochar prepared by the hydrothermal processing of selected feedstock.<sup>a</sup>

Feedstock	HT	Reaction conditions			Yield (wt%)	Proximate analysis			Ultimate analysis (wt%)					Atomic ratio		pH	HHV (MJ/kg)	Ref.
		T (°C)	RT (h)	F/W		FC	VM	Ash	C	H	O	N	S	H/C	O/C			
Loblolly pine	HTC	200-230	0.01-7-0.083	1:30	62.44-79.09	14.4-4-17.04	82.9-1-86.13	-	51.4-4-53.05	6.14-6.48	40.3-9-42.17	-	-	-	-	-	21.41-22.39	<sup>1</sup>
Sewage sludge	HTC	180-260	0.5-8	1:9	53.00-66.19	6.14-13.24	16.7-9-34.33	59.5-3-72.02	19.6-2-24.49	3.22-3.73	2.13-10.54	1.12-1.64	0.49-0.67	1.80-1.99	0.07-0.33	-	9.80-11.79	<sup>2</sup>
Sludge	HTC	220	5	1:3-4:1	55.12-63.96	10.6-3-20.74	25.0-6-34.52	53.5-3-59.83	27.3-7-29.03	3.90-4.25	0.55-7.67	5.35-5.81	0.52-0.60	-	-	-	10.65-12.89	<sup>3</sup>
Sewage sludge	HTC	160-280	-	-	65.64-77.50	1.89-4.95	12.7-4-26.02	72.0-9-82.31	9.99-13.65	1.08-1.77	4.39-9.46	1.53-2.30	0.58-0.73	1.28-1.56	0.33-0.52	-	-	<sup>4</sup>
Sewage sludge (10% humic acid)	HTC	250	5	-	41.3	-	30.61	60.39	16.84	2.45	9.14	1.88	0.3	-	-	-	7.66	<sup>5</sup>
Beet pulp	HTC	180-220	1-4	-	-	20.1-29.46	77.7-66.71	0.88-1.00	48.0-60.8	5.67-6.15	26.3-0-40.33	1.61-2.09	0-0.1	-	-	3.9-4-4.45	19.61-25.36	<sup>6</sup>
Rice straw	HTC	180-260	4	8:25	-	-	-	18.1-7-24.41	43.5-1-52.75	4.40-4.75	16.8-5-32.42	1.16-1.60	-	0.27-0.36	0.40-0.92	-	-	<sup>7</sup>
Digestate	HTC	210	0.5-5	-	70.0-75.6	20.1-22.4	45.6-49.9	29.6-31.2	41.9-42.4	3.4-3.8	19.8-22.0	2.3-2.4	0.26-0.28	-	-	-	16.0-16.3	<sup>8</sup>
Rape straw	HTC	160-240	0.5	-	-	12.8-6-32.51	56.6-6-79.90	7.24-10.83	44.9-0-65.67	4.98-5.68	15.1-7-38.77	2.23-2.91	0.78-1.33	0.91-1.48	0.17-0.65	-	18.13-27.04	<sup>9</sup>

		180	0.25- 2	-	-	12.7 9- 21.1 6	71.3 8- 79.1 7	7.46- 8.04	53.3 9- 58.4 4	5.24- 5.47	24.9 2- 29.5 0	2.49- 2.59	1.16- 1.34	1.09- 1.18	0.32- 0.41		19.88-22.83		
Cellulose	HTC	150 – 250	0.5	–	50.4 – 92.5	9.6 – 41.2	56.9 – 88.9	1.2 – 1.9	43.9 – 69.4	4.6 – 6.6	24.1 – 48	–	–	0.80 – 1.80	0.26 – 0.82	–	16.6 – 26.8	<sup>10</sup>	
Holocellulose	HTC	220	4 – 20	3:20	22.0 – 66.7	5.58 – 50.4	44.2 – 87.9	1.31 – 5.10	48.7 – 73.0	4.32 – 5.71	22.6 – 45.6	–	–	0.71 – 1.60	0.23 – 0.85	–	16.5 – 26.8	<sup>11</sup>	
Bamboo	HTC	200 – 260	1	1:10	39.3 – 64.8	1.09 – 43.9	55.1 – 98.0	0.95 – 1.11	56.0 – 73.2	5.52 – 5.99	21.3 – 37.6	0.42 – 0.69	0.02 – 0.03	0.91 – 1.28	0.22 – 0.50	5.2 – 5.4	–		<sup>12</sup>
Tomato peel	HTC	150 – 250	1.6 – 18.4	2:125- 3:20	27.6 – 87.7	–	–	–	51.9 – 69.7	7.2 – 8.2	20.9 – 39.4	0.58 – 2.00	0.05 – 0.65	1.31 – 1.70	0.23 – 0.57	–	23.6 – 34.8	<sup>13</sup>	
Corn husk	HTC	260	0.25	1:12	33.2	28.4	68.1	3.7	63.4	5.87	25.1	1.93	<0.01	1.11	0.30	–	27.7	<sup>14</sup>	
Corn cobs	HTC	230 – 260	0.5	1:6	–	–	–	–	59.0 – 66.9	5.18 – 5.72	27.0 – 34.5	0.67 – 0.87	0.10 – 0.12	0.93 – 1.17	0.30 – 0.40	–	21.9 – 25.2	<sup>15</sup>	
Wheat straw	HTC	200 – 260	6	1:20	35.7 – 53.5	–	–	9.3 – 10.8	51.2 – 64.2	4.8 – 5.7	18.8 – 33.1	0.5 – 1.1	0.2 – 0.3	0.90 – 1.34	0.22 – 0.48	4.9 – 6.3	21.0 – 26.2	<sup>16</sup>	
Grape pomace	HTC	175 – 275	0.17 –1	1:4	46.5 – 61.1	29.2 – 43.0	54.3 – 69.3	1.1 – 3.4	58.6 – 68.3	5.8 – 6.4	23.5 – 33.0	1.72 – 2.32	–	1.04 – 1.27	0.26 – 0.42	–	24.3 – 28.3	<sup>17</sup>	
Miscanthus	HTC	180 – 200	2 – 12	1:10	58 – 80	15.5 – 24.3	73.2 – 81.7	0.59 – 1.79	53.5 – 63.0	5.8 – 6.2	29.9 – 39.2	0.07 – 0.29	–	1.10 – 1.33	0.36 – 0.55	–	21.1 – 23.9	<sup>18</sup>	
Cotton stalk	HTC	180 – 260	1 – 8	1:15	~34.0 – 61.8	15.0 – 43.4	47.4 – 71.5	5.16 – 12.0	51.2 – 70.6	4.76 – 5.81	10.3 – 36.6	0.68 – 1.57	–	0.91 – 1.31	0.11 – 0.54	–	19.7 – 25.1	<sup>19</sup>	
Solid digestate	HTC	180 – 260	1	1:8	46.3 – 80.7	–	–	9.60 – 16.7	55.2 – 69.8	6.42 – 7.08	19.8 – 35.2	1.88 – 3.22	0.71 – 0.78	1.09 – 1.53	0.21 – 0.48	5.8 – 6.8	24.1 – 29.9	<sup>20</sup>	
Lipid extracted algae	HTC	180 – 240	0.5	–	51.8 – 74.5	10.4 –	78.3 –	4.10 –	54.5 –	7.33 –	21.8 –	8.10 –	0.34 – 0.42	1.54 –	0.27 –	–	23.6 – 27.7	<sup>21</sup>	

					16.3	85.5	6.54	59.7	8.04	27.5	8.94		1.73	0.38				
Sawdust	HTC	220	1	1:5	56.5	18.8	76.6	1.60	50.6	5.35	41.8	0.59	0.09	1.26	0.62	–	20.5	22
Sewage sludge					59.1	4.40	35.8	59.8	23.6	2.84	10.6	2.56	0.56	1.43	0.34	–	11.8	
Sewage sludge	HTC	220	1	1:9	–	15.7	31.7	52.6	43.6	4.79	42.6	9.52	1.03	1.32	0.73	–	16.2	23
Sawdust	HTC	220	10	1:6	68.2	24.6	74.4	0.85	58.1	6.06	34.1	0.91	0.01	1.25	0.44	–	23.9	24
Swine manure					48.7	10.3	69.8	20.4	49.5	5.78	20.8	3.15	0.35	1.40	0.32	–	21.5	
Mixture <sup>b</sup>					60.5	16.4	71.9	11.7	51.0	5.92	28.1	2.94	0.33	1.39	0.41	–	21.62	
Cow manure	HTC	250-300	0.5-2	1:10	51.29-42.25	26.5-3-54.1	30.2-3-46.0	27.4-3-34.2	46.5-8-54.1	3.76-4.29	20.2-8-5.88	1.31-1.71	0.11-0.32	0.81-1.10	0.08-0.33	–	19.52-23.62	25
Swine manure	HTC	180–200	10	1:4	55.5–66.4	5.57–8.06	68.7–73.1	21.3–24.8	46.2–50.6	5.44–5.72	17.1–23.1	3.00–3.24	0.40–0.45	1.29	0.25	–	–	26
Corn stalk <sup>c</sup>	HTC	230	0.75	1:25	39.0	18.0	74.3	3.54	53.4	5.67	39.6	1.12	0.13	1.27	0.56	–	22.8	27
		190	0.5	7:100	64.7	11.6	80.7	3.12	48.9	5.85	43.9	1.14	0.17	1.43	0.67	–	21.7	
Red seaweed <sup>c</sup>	HTC	180	0.33	3:100	32.1	24.4	59.9	15.7	45.6	6.0	46.6	1.9	–	1.58	0.77	–	17.9	28
Cellulose	HTL	280	0.5	–	46.2	55.1	42.2	2.7	76.5	4.5	16.3	–	–	0.71	0.16	–	27.7	10
Cellulose	HTL	280	4	1:10	34.0	–	–	–	72.9	4.73	22.4	–	–	0.78	0.23	–	27.1	29
Xylan					–	–	–	–	73.0	4.65	22.6	–	–	0.76	0.23	–	28.1	
Bamboo	HTL	280	1	1:10	36.7	35.5	63.8	0.71	74.6	5.37	19.2	0.81	0.03	0.86	0.19	5.3	–	12
Coconut fiber	HTL	300–350	0.5	1:10	~ 35–40	38.5–42.1	53.6–56.6	4.3–4.9	73.2–73.4	4.52–5.09	20.2–20.6	1.13–1.17	0.35–0.36	0.74	0.21	–	28.7–29.4	30
Eucalyptus leaves					~ 30–40	31.7–33.9	56.2–61.2	7.1–9.9	68.9–70.5	5.93–6.00	20.5–22.8	1.60–1.62	0.72–1.52	1.00	0.22	–	28.7–29.4	
Tahoe mix	HTL	295	0.5	1:8	50.1	–	–	–	73.0	5.14	19.9	0.14	–	–	0.20	–	29.5	31, 32
Loblolly pine					52.4	–	–	–	72.1	4.91	21.1	0.36	–	–	0.22	–	29.0	
Pinyon/Juniper					48.7	–	–	–	71.1	5.25	21.6	0.34	–	–	0.23	–	28.0	
Sugarcane bagasse					42.8	–	–	–	64.8	4.69	19.6	0.72	–	–	0.23	–	24.2	
Corn stover					38.0	–	–	–	67.4	4.78	18.1	1.18	–	–	0.20	–	26.3	
Rice hulls					52.2	–	–	–	49.7	3.35	15.5	0.36	–	–	0.23	–	18.5	
Cotton stalk	HTL	280–300	4	1:15	~ 25.8–28	39.8–	45.8–	3.21–	72.1–	5.08–	9.22–	1.59–	–	0.82	0.09	–	25.5–25.7	19

					43.1	50.1	9.64	73.9	5.28	17.1	1.81		0.88	0.18					
Sewage sludge/wheat stover	HTL	350	0.25-1.5	1:10	36.66-56.51	-	-	-	17.8	7-	1.56-4.04	2.49-7.32	1.15-1.90	0.35-0.49	1.04-1.5	0.09-0.27	-	7.53-15.88	33
Cow manure	HTL	400	0.2-0.5	1:10	38.90-11.31	-	-	-	49.1	3.38-6-	36.7	0.87-1.61	0.55-0.59	0.70-1.23	0.48-0.68	-	18.51-20.43	34	
Cow manure	HTL	425	0.2-0.33	1:10	12.08-45.13	-	-	-	41.0	2.70-4.86	47.0	0.98-1.72	0.66-0.84	0.79-1.32	0.76-0.99	-	11.07-16.55	35	
Rice straw	HTL	300	1.5	1:20	25.6	26.4	52.2	-	-	-	-	-	-	0.94	0.17	5.9	-	36	
Pig manure				3:20	43.7	35.4	26.8	-	-	-	-	-	-	0.91	0.19	6	-		
Sugarcane bagasse	HTL	300	6	4:31	26.8	-	-	-	79.3	5.34	15.4	0.88	0.11	0.07	0.19	5.8	-	37	
Hickory				5:29	27.8	-	-	-	78.5	5.14	16.4	0.33	0.06	0.07	0.21	5.4	-		
Peanut hull				11:62	36.9	-	-	-	76.4	6.07	17.5	2.06	0.13	0.08	0.23	6.0	-		
Eucalyptus bark	HTL	300	2	1:10	40.0	34.0	60.1	5.9	72.7	5.05	22.2	-	-	0.83	0.23	-	29.2	38	
Sawdust	HTL	300	1	1:10	24.9	-	-	0.52	-	-	-	-	-	1.02	0.28	4.1	-	39	
<i>Salix psammophila</i>	HTL	300	1	2:25	-	-	-	15.2	66.3	5.21	27.2	1.26	-	0.94	0.31	5.2	-	40, 41	
<i>Salix psammophila</i>	HTL	300	1	1:10	35.0	-	-	7.92	73.6	7.90	17.7	0.80	-	1.29	0.18	-	-	42	
<i>S. platensis</i>	HTL	350	3	1:3	6.0	-	-	-	24.6	2.61	6.62	2.07	0.30	0.23	-	13.1	-	43	
<i>N. salina</i>	HTL	310-350	2-3	1:3	28.0-32.0	-	-	-	67.7-67.9	7.26-7.75	9.30-10.6	2.63-2.65	0.93-1.76	0.41-0.47	-	-	32.5-33.6	43	
Coconut fiber	HTG	375	0.5	1:10	35.3	48.8	42.6	8.6	78.2	4.31	15.9	1.23	0.33	0.66	0.15	-	30.6	30	
Eucalyptus leaves					28.1	42.6	43.2	14.2	72.2	4.81	19.9	1.64	1.51	0.80	0.21	-	28.7		
Wheat straw	HTG	460	0.33	1:100	-	-	-	9.8	66.9	2.05	18.2	2.4	0.65	0.37	0.20	-	-	44	
<i>C. glomerata</i>					-	-	-	36.2	44.2	2.5	10.6	3.4	3.1	0.68	0.18	-	-		

<sup>a</sup> HT: hydrothermal technologies; T: temperature; RT: retention time; F/W: feed to water mass ratio; FC: fixed carbon; VM: volatile matter; HHV: higher heating value.

<sup>b</sup> Hydrochar was produced from co-HTC of swine manure and sawdust in the mass ratio of 3:1.

<sup>c</sup> Hydrochar was produced from microwave-assisted HTC.

**Table S2** Main inorganic element content in pristine hydrochar obtained from hydrothermal processing of various feedstocks.

Feedstock	HT	Reaction conditions			P (g/kg)	K (g/kg)	Ca (g/kg)	Mg (g/kg)	Na (g/kg)	Si (g/kg)	Al (g/kg)	Fe (g/kg)	Cu (g/kg)	Zn (g/kg)	Ni (g/kg)	Ti (g/kg)	Ref.
		T (°C)	RT (h)	F/W													
Sewage sludge	HTC	180	1	–	–	7.2	14.4	18.9	3.3	18.5	21.8	20.3	2.8	1.5	2.0	1.4	45
Sewage sludge	HTC	220	1	1:5	–	18.1	17.4	12.3	6.7	17.4	35.0	43.4	1.9	0.7	0.4	13.7	22
Sawdust					–	2.5	9.0	1.6	1.1	2.6	0.8	1.0	0.3	0	0	1.7	
Mixture <sup>c</sup>					–	5.4 – 10.9	5.0 – 13.0	3.2 – 9.0	2.0 – 4.3	5.0 – 10.4	10.3 – 21.4	10.3 – 31.7	0.8 – 1.4	0.2 – 0.5	0 – 0.2	3.4 – 9.7	
Lincomycin residue <sup>d</sup>	HTC	120 – 210	1	1:10	–	–	87.5 – 167	1.08 – 2.96	–	–	1.91 – 2.36	11.5 – 23.2	–	– <sup>r</sup>	–	–	46
Cow manure	HTC	275	30	1:10	–	0.22	9.78	5.68	0.12	–	0.48	2.01	–	–	–	–	25
Herbal tea waste	HTC	120 – 240	0.5	1:10	3.35 – 8.45	6.61 – 9.62	43.11 – 47.60	3.79 – 6.23	0.51 – 0.64	28.39 – 40.06	10.01 – 15.06	12.09	–	–	–	1.31 – 2.37	47
	HTL	300			4.68	6.75	49.98	20.39	0.40	78.95	26.99	46.81	–	–	–	3.93	
Penicillin mycelial waste <sup>d</sup>	HTC	120 – 240			16.15 – 65.12	4.22 – 11.07	57.26 – 163.13	2.59 – 11.60	0.60 – 1.06	0.69 – 1.27	0.86 – 1.70	1.87 – 6.78	–	–	–	–	
	HTL	300			113.39	4.16	232.97	21.58	0.61	2.02	2.04	8.36	–	–	–	–	
Sewage sludge	HTC	120 – 240			33.0 – 39.88	16.29 – 17.73	21.01 – 23.50	11.63 – 14.66	1.34 – 1.42	190.53 – 200.31	87.44 – 106.51	42.17	–	–	–	4.84 – 5.06	
	HTL	300			39.05	18.62	24.14	15.56	1.39	210.61	109.78	46.48	–	–	–	5.10	
Swine manure	HTL	330	0.5	1:5	30000	4050	13261	13980	1757	–	18750	16902	3940	2930	64	–	48
Micoralgae					80569	12590	29146	2001	4826	–	9001	20015	425	900	189	–	
Salix psammophila	HTC	180 – 270	0 – 4	1:10	–	0.12 – 0.13	10.9 – 13.2	0.16 – 0.33	0.02 – 0.04	–	–	–	–	–	–	–	49

	HTL	300	1	1:10	–	0.15	7.38	0.64	0.02	–	–	–	–	–	–	–	
Rice straw	HTL	300	1	1:10	–	0.42	5.63	3.52	0.01	–	–	–	–	–	–	–	
Bamboo					–	0.01	BDL	0.03	BDL	–	–	–	–	–	–	–	
Soybean straw					–	0.43	3.21	0.32	BDL	–	–	–	–	–	–	–	
Pomelo peel					–	0.09	2.97	0.30	BDL	–	–	–	–	–	–	–	
Pine needle					–	0.29	7.05	0.86	0.04	–	–	–	–	–	–	–	
<i>Enteromorpha prolifera</i>					–	1.89	47.1	12.7	0.24	–	–	–	–	–	–	–	
Sugarcane bagasse	HTC	200- 250	6	4:31	0 – 0.2	0.1	0.3 – 0.4	0.2 – 0.3	0.3	–	0.1	0.1	–	–	–	–	37
	HTL	300			0.3	0.1	0.6	0.3	0.4	–	0.1	0.1	–	–	–	–	
Hickory	HTC	200- 250		5:29	0	0.1	0.7 – 0.8	0.3 – 0.4	0.2 – 0.4	–	0.1	0.1 – 0.3	–	–	–	–	
	HTL	300			0.1	0.1	1.3	0.1	0.1	–	0.1	0.4	–	–	–	–	
Peanut hull	HTC	200- 250		11:62	0.6 –	0.3	0.7 –	0.2 –	0.2 –	–	0.1 –	0.2 –	–	–	–	–	
	HTL	300			0.7		0.8	0.3	0.3		0.4	0.4					
	HTL	300			1.7	0.3	1.6	0.5	0.2	–	0.4	0.3	–	–	–	–	
Wheat straw	HTG	460	0.33	1:100	–	41.1	54.2	5.7	18.0	–	–	–	–	–	–	–	44
<i>C. glomerata</i>					–	294	122	82	175	–	–	–	–	–	–	–	
Cattle manure	HTG	440	0.33	1:39	–	57.4	48.6	13.4	26.7	–	–	–	–	–	–	–	50

<sup>a</sup> HT: hydrothermal technologies; T: temperature; RT: retention time; F/W: feed to water mass ratio; BDL: below detection limit;

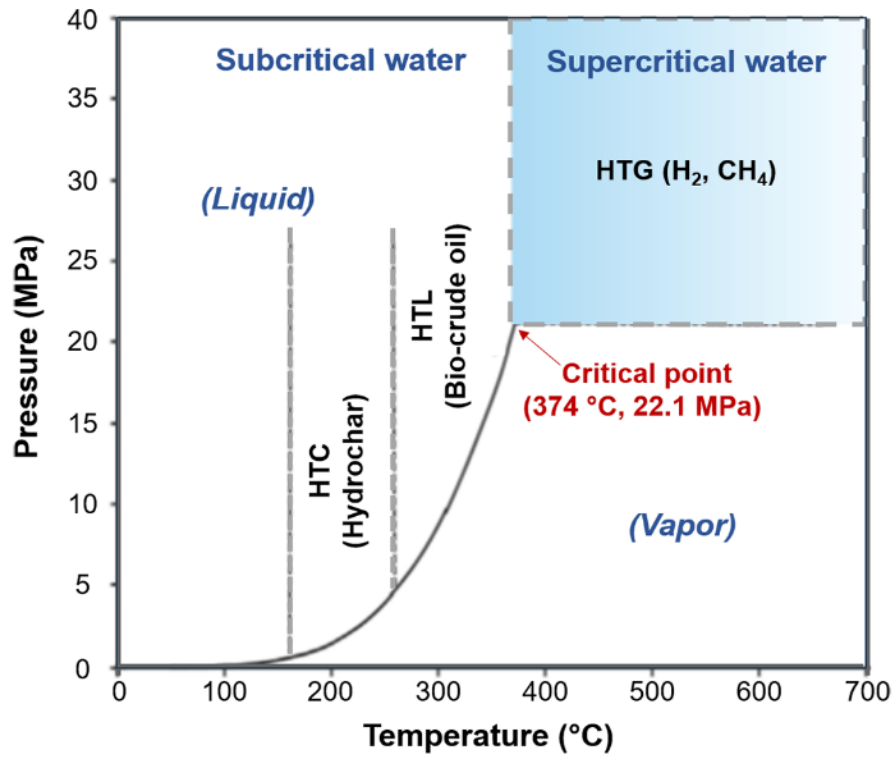
<sup>b</sup> Hydrochar was produced from co-HTC of swine manure and sawdust in the mass ratio of 3:1 with water recirculation from 0 – 4 times;

<sup>c</sup> Hydrochar was produced from co-HTC of sawdust and sewage sludge in the mass ratio of 1:3 – 3:1.

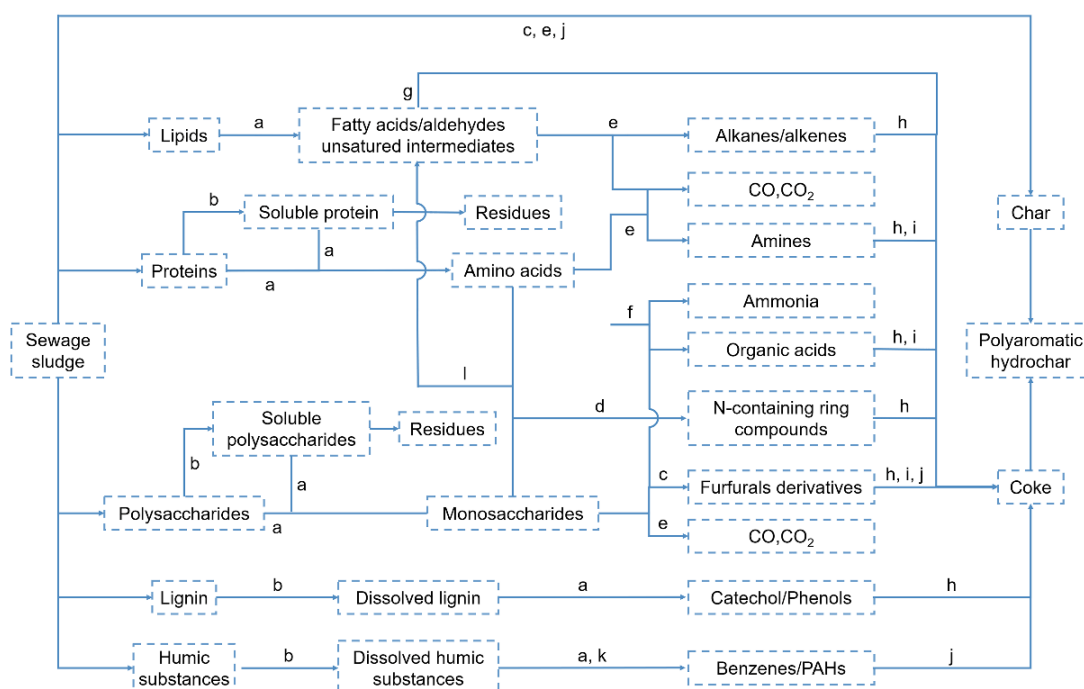
<sup>d</sup> Hydrochar was produced from microwave-assisted HTC.







**Fig. S1** Classification of hydrothermal regions and their resulting principles products with reference to the pressure-temperature phase diagram of water. Adapted from ref.<sup>51-54</sup>



**Fig. S2** Schematic reaction pathways of sewage sludge to hydrochar: (a) hydrolysis, (b) solubilization, (c) dehydration, (d) Maillard reaction, (e) decarboxylation, (f) deamination, (g) aggregation, (h) polymerization, (i) aromatization, (j) condensation, (k) decomposition, (l) C-C cleavage, and PAHs=polycyclic aromatic hydrocarbons. Adapted from ref.<sup>55</sup>

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