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Outlook and challenges for recovering energy and water from complex organic wastes using hydrothermal liquefaction

Daniela Cabrera, Rodrigo Labatut*

Department of Hydraulic and Environmental Engineering, Pontificia Universidad Católica de Chile, Santiago, Chile.

***Corresponding author.**

Tel.: +56-9-79312858; Email: rolabatut@ing.puc.cl

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Table S1. Ultimate analysis (%), yields (wt%), and higher heating value - HHV (MJ/kg) of upgraded bio-crude.

HTL Feedstock	Biocrude	Process	Catalyst	P H_2 (MPa)	Temp (°C)	Time (min)	Better conditions	Yield (wt.%)	C	H	N	S	O	HHV	Ref.
	Northern sea crude								86.6	13.1	n.d.	n.d.	0.3	44.4	[54]
Chlorella	RB	-	-	-	-	-	-	-	72.8	9.4	6.0	0.8	11.1	36.1	[55]
	UB	Dry	CoMo, NiMo, No cat.	3.8	350-405	120	CoMo, 3.8 MPa H_2, 405 °C	69.4	84.4	11.9	2.7	0.0	1.0	45.4	
Chlorella pyrenoidosa	RB	-	-	-	-	-	-	-	76.4	10.4	7.7	0.76	5.5	39.8	[56]
	UB	Wet	Ru/C with Pd/C, Pt/C, Pt/C-S, Pt/γ-Al_2O_3, Rh/γ- Al_2O_3, CoMo/ γ-Al_2O_3-S, MoS_2, Mo_2C, Raney-Ni, activated carbon, or alumina; No cat.	6	400	240	Ru/C + Mo_2C	77.2	83.9	12.9	3.1	0.08	0.1	46.8	
Chlorella pyrenoidosa	RB	-	-	-	-	-	-	-	79.2	10.8	8.0	NR	2.1	39.8	[57]
	UB	Wet	Ru/C, Pd/C, Pt/C, Pt/C-S, Pt/C(CO), Pt/C(n-C_6H_{14}), Mo_2C, Ni/SiO_2- Al_2O_3, CoMo/Al_2O_3, HZSM-5, MoS_2, Raney-Ni, activated carbon,	6	400	240	Ru/C + Raney-Ni	77.2	83.9	12.1	2.0	NR	2.0	45.3	

			alumina, Ru/C + Raney-Ni, No cat.												
Nannochloropsis sp.	RB	-	-	-	-	-	-	-	77.3	10.5	4.8	0.7	6.5	40.1	[58]
	UB	Wet	Pt/C, No cat.	0 - 3.4	400	240	Pt/C, 3.4 MPa H_2	82[a]	82.1	11.2	2.2	n.d.	4.5	43.0	
Nannochloropsis sp.	RB	-	-	-	-	-	-	-	75.9	10.4	4.8	0.5	8.1	39.2	[59]
	UB	Wet	Pt/C, Mo_2C, HZSM-5	3.5	430-530	120-360	HZSM-5, 430 °C, 360 min	76[a]	84.4	10.8	2.4	n.d.	2.5	43.5	
Nannochloropsis gaditana	RB	-	-	-	-	-	-	-	74.4	10.1	4.8	0.5	10.3	37.0	[60]
	UB	Dry, wet	Pt/Al_2O_3, HZSM5, No cat.	4 - 8	400	240	Dry, Pt/Al_2O_3, 8 MPa H_2	61.9	84.2	11.7	2.4	<0.1	1.6	43.2	
Scenedesmus almeriensis	RB	-	-	-	-	-	-	-	74.2	9.4	5.7	0.8	10.0	36.3	[60]
	UB	Dry, wet	Pt/Al_2O_3, HZSM5, No cat.	4 - 8	400	240	Dry, Pt/Al_2O_3, 8 MPa H_2	61.8	82.7	11.0	4.2	0.2	1.9	41.9	
Spirulina	RB	-	-	-	-	-	-	-	75	10.4	7.7	NR	6.9	37.7	[61]
	UB	Dry	$NiMo/Al_2O_3$	4 - 8	350-400	240	8 MPa H_2, 350 °C	73	84	12.1	4.0	NR	0.0	43.5	
Hard wood	RB	-	-	-	-	-	-	-	83.9	10.4	0.4	n.d.	5.3	40.43	[54]
	UB	Dry	$NiMo/Al_2O_3$	152-	150-	120	350 °C, 337 NL H_2/L bio-	-	87.2	12.2	0.4	n.d.	0.3	43.90	

				550[b]	350		crude								
Miscanthus	RB	-	-	-	-	-	-	-	70.5	8.2	1.7	NR	19.6	32.2	[61]
	UB	Dry	$NiMo/Al_2O_3$	4 - 8	350-400	240	8 MPa H_2, 400 °C	61	87.4	10.3	1.5	NR	0.8	42.2	
Duckweed	RB	-	-	-	-	-	-	-	73.4	7.9	4.7	0.38	13.6	33.5	[62]
	UB	Wet	Ru/C, Pd/C, Pt/C, Pt/C (sulfide), Pt/γ-Al_2O_3, Rh/γ- Al_2O_3, CoMo/ γ-Al_2O_3 (sulfide), MoS_2, Mo_2C, activated carbon, zeolite, No cat.	6	350	240	Ru/C	67.6	83.6	11.5	4.5	0.13	0.3	42.6	
Primary sludge from WWTP	RB	-	-	-	-	-	-	-	74.5	10.6	3.9	NR	11.0	37.4	[61]
	UB	Dry	$NiMo/Al_2O_3$	4 - 8	350-400	240	8 MPa H_2, 400 °C	72	85.3	13.8	0.9	NR	0.0	46.1	
Primary sludge from WWTP	RB	-	-	-	-	-	-	-	76.5	10.1	4.3	0.63	8.43	38.8[c]	[41]
	UB	Dry	CoMo-S	1276[b]	400	NR	-	76.7	84.7	14.2	0.03	22[d]	1.1	48.7[c]	
Anaerobic digestate from WWTP	RB	-	-	-	-	-	-	-	82.3	9.3	4.7	1.14	2.6	40,6[c]	[41]
	UB	Dry	CoMo-S	1276[b]	400	NR	-	93.1	85.0	14.2	0.06	24[d]	0.96	48,8[c]	

RB: Raw biocrude; UB: Upgraded biocrude; NR: Not reported; n.d.: Not detected; WWTP: Waste water treatment plant.

[a] Carbon basis

[b] Normal litre hydrogen per litre bio-crude (NL/L)

[c] Calculated according to the Dulong's formula [53]: HHV (MJ/kg) = 0.338C + 1.428(H - O/8)

[d] Expressed as parts per million (ppm)

Table S2. Operating conditions of HTL and main results for selected organic wastes

Feedstock	Type of reactor	Process gas / Catalyst	Temp (°C)	Reaction time (min)	Volume of slurry fed (mL)	Slurry TS (wt%)	Slurry COD (g COD/L) [HHV (MJ/kg)] [a]	Biocrude yield (wt%) [HHV (MJ/kg)] [a]	AP yield (wt%)[b] [COD (g COD/L)]	Hydrochar yield (wt%)	Ref.
Pre-digested WAS	Continuous, 500 mL	N_2 / No cat.	350	19	1500 [c]	9.7	153 [16.2]	24.8 [35.8]	15.8 [d] [73]	1.99	41
Post-digested PS+WAS	Continuous, 500 mL	N_2 / No cat.	350	30	1500 [c]	16	203 [18.9]	34.4 [39.0]	16.8 [d] [48.2]	5.7	41
Swine manure	Batch, 1800 mL	N_2 / No cat.	305	120	800 [e]	20	237.4 [e] [19.1] [e]	48.1 [36.6] [f]	14.0 [e] [77.4]	3.3	17
Food waste	Batch, 500 mL	N_2 / No cat.	300	40	200	5	48 [19.7]	28.7 [35.1]	27.6 [7.3] [g]	8.9 [h]	63
Sorghum bagasse	Batch, 1000 mL	N_2 / K_2CO_3	300	60	100	28	178.2 [i] [15.5] [j]	45.3 [29.7]	- [k] [23.1] [j]	18.35	64

TS: Total solid content; COD: chemical oxygen demand; AP: aqueous product; HHV: higher heating value; WAS: waste activated sludge; PS: primary sludge

[a] Calculated according to the Dulong's formula [53]: HHV (MJ/kg) = 0.338C + 1.428(H - O/8)

[b] The volume of aqueous product was assumed as equal to the volume of water supplied to the HTL reactor

[c] Feed rate (mL/h)

[d] Value from [65]

[e] Value from [66]

[f] Value from [67]

[g] Value from [68]

[h] Value from [37]

[i] Calculated according to McCarty (1964) [69]

[j] Value from [5]

[k] Total solid content of the aqueous product was not reported

Table S3. MM dry tons of waste annually produced, and excess currently not being used in selected regions.

Feedstock	US	US Excess	EU	EU Excess	China	China Excess
Wastewater sludge	· 14.82 [70 h]	7.7 [70 h]	11.1 [a 71]	6.2 [a,b 71,72]	40 [d 73]	32 [d 73]
Animal manure	41 [70]	26 [70]	122 [f 74]	33.4 [f 74]	554 [c 75]	152.9 [c 75]
Food waste	15.3 [70 c]	14 [70 c]	87.6 [g 76]	78.4 [g 76]	202.5 [g 77]	114.6 [g 77]
Agricultural waste	524 [78]	17.1 [78]	91.6 [f 74]	72.8 [f 74]	767 [c 75]	240.1 [c 75]

[a] Data from 2013

[b] Percentage of used sludge is estimated from Eurostat 2012 [72], considering agricultural use, compost and other.

[c] Data from 2012

[d] Data from 2014

[e] Data from 2015

[f] Average value over the years 2009-2011 from EU

[g] Data from 2011

[h] Data from 2016

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