Electronic Supplementary Information

Techno-economic analysis and life cycle assessment of mixed plastic waste gasification for production of methanol and hydrogen

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gasification | mixed plastic waste | methanol | hydrogen | TEA | LCA | municipal solid waste

S1. Supporting Figures

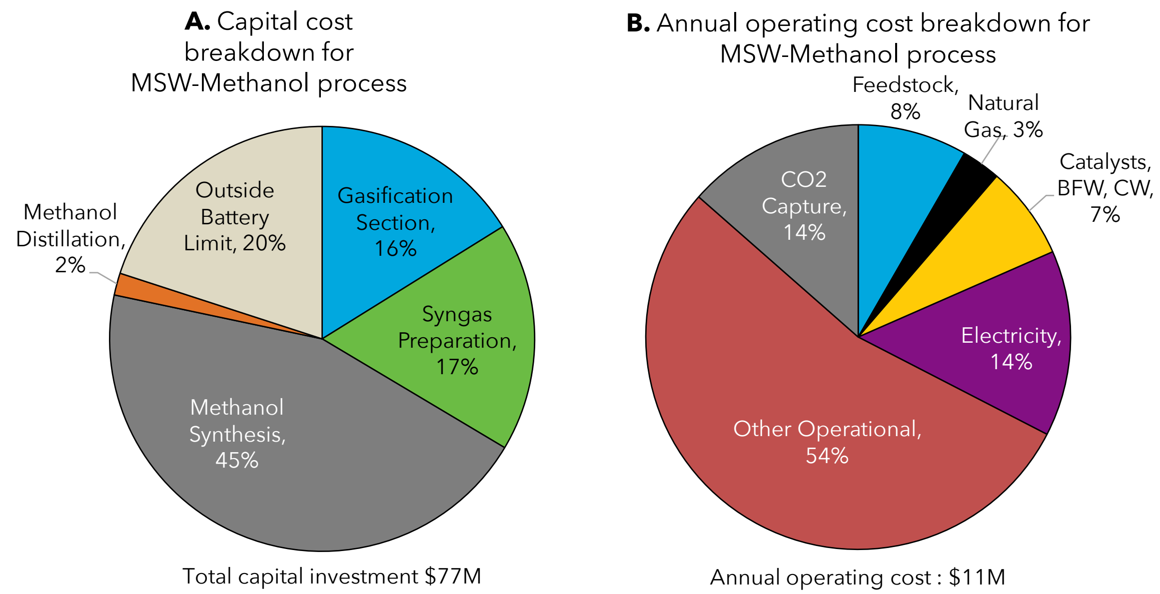

Fig. S1. *CAPEX and OPEX breakdown for MSW-Methanol process*

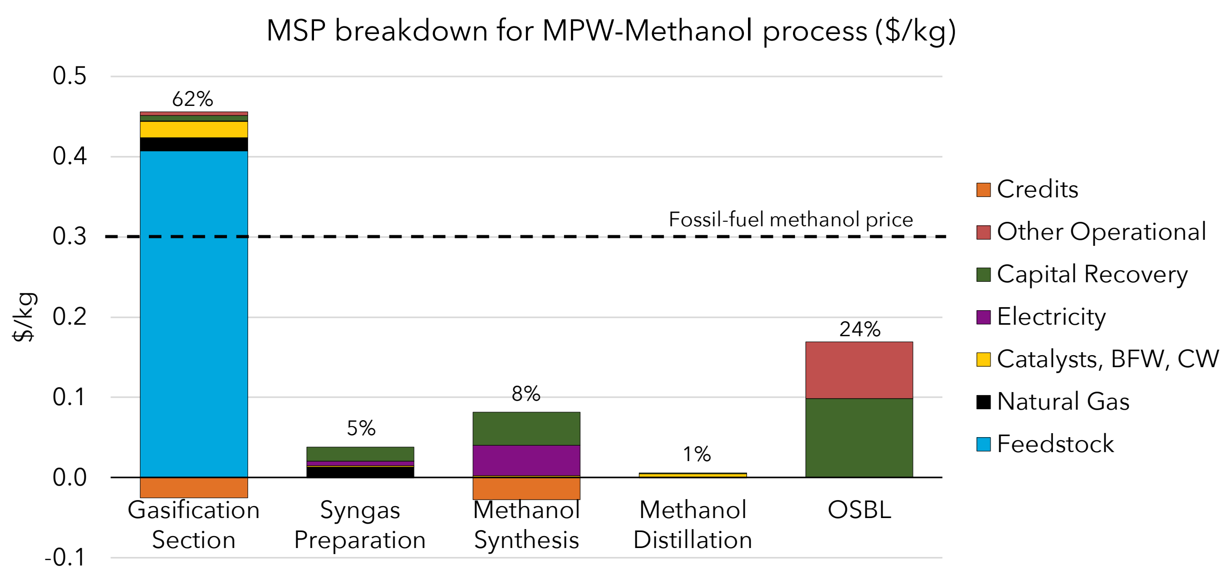

Fig. S2. *Methanol MSP breakdown by process area for MPW-Methanol process*

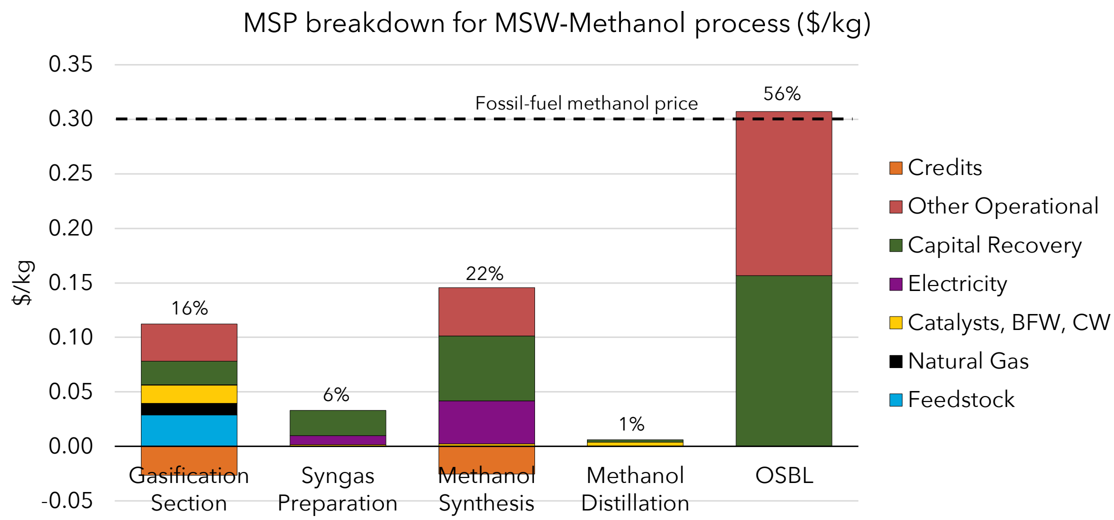

Fig. S3. *Methanol MSP breakdown for MSW-Methanol process*

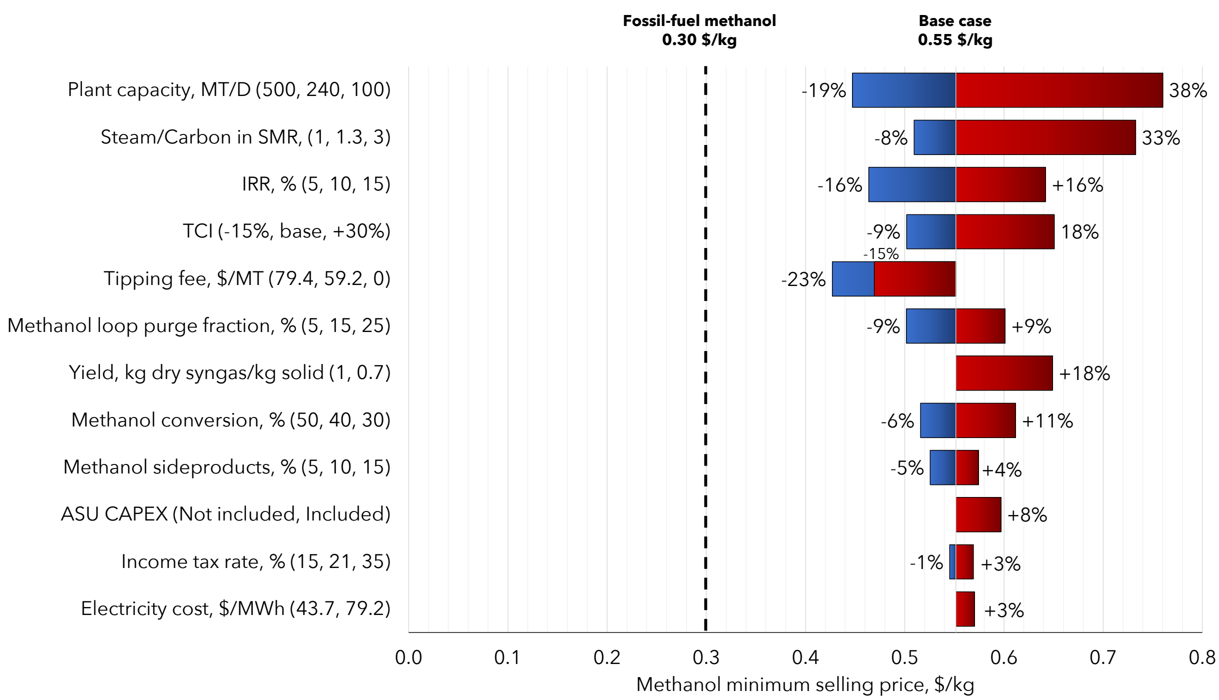

Fig. S4. *Sensitivity analysis for MSW-Methanol process.* Economies of scale effect is more apparent in MSW-Methanol process. Considering a credit based on the US average tipping fee of $59/MT, the methanol MSP is $0.49/kg. Discussion on these results is included in section **S4**.

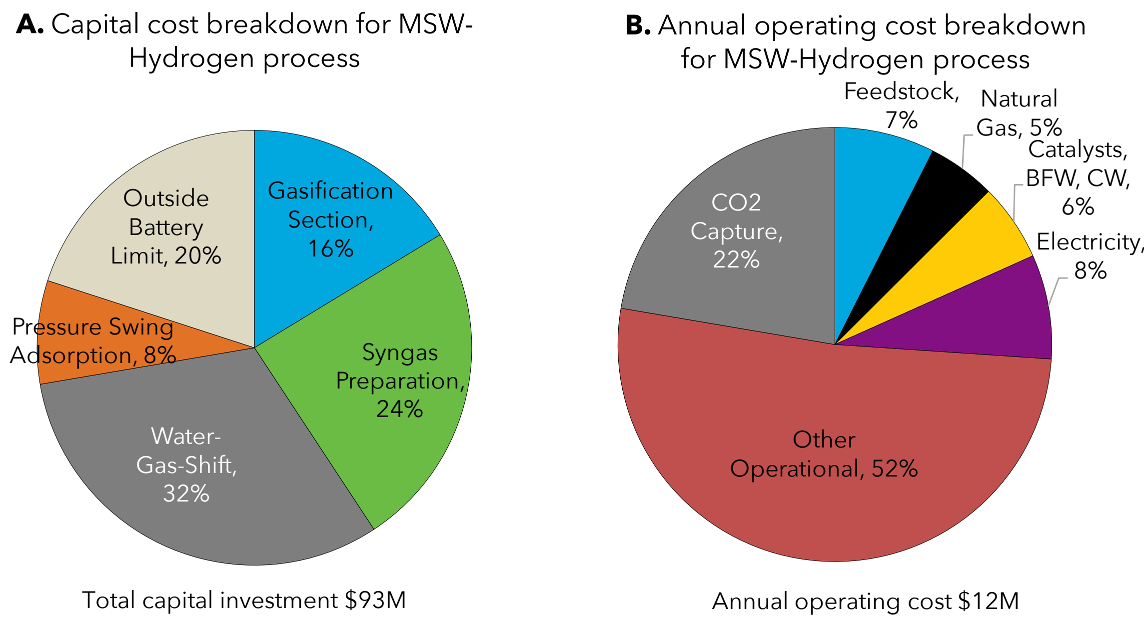

Fig. S5. *CAPEX and OPEX breakdown for MSW-Hydrogen process*

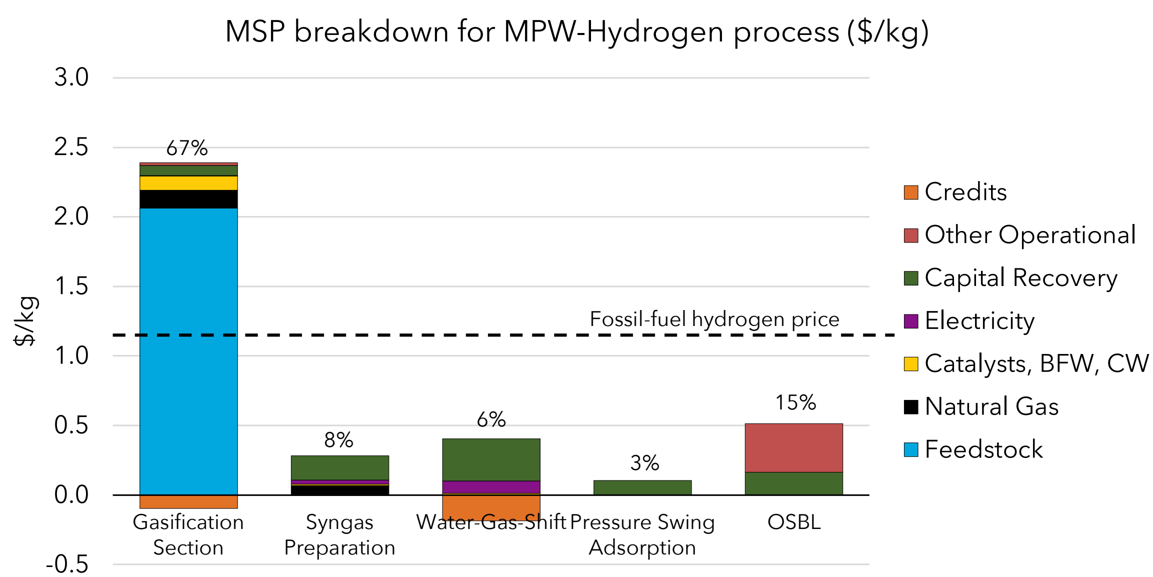

Fig. S6. *Hydrogen MSP breakdown by process area for MPW-Hydrogen process*

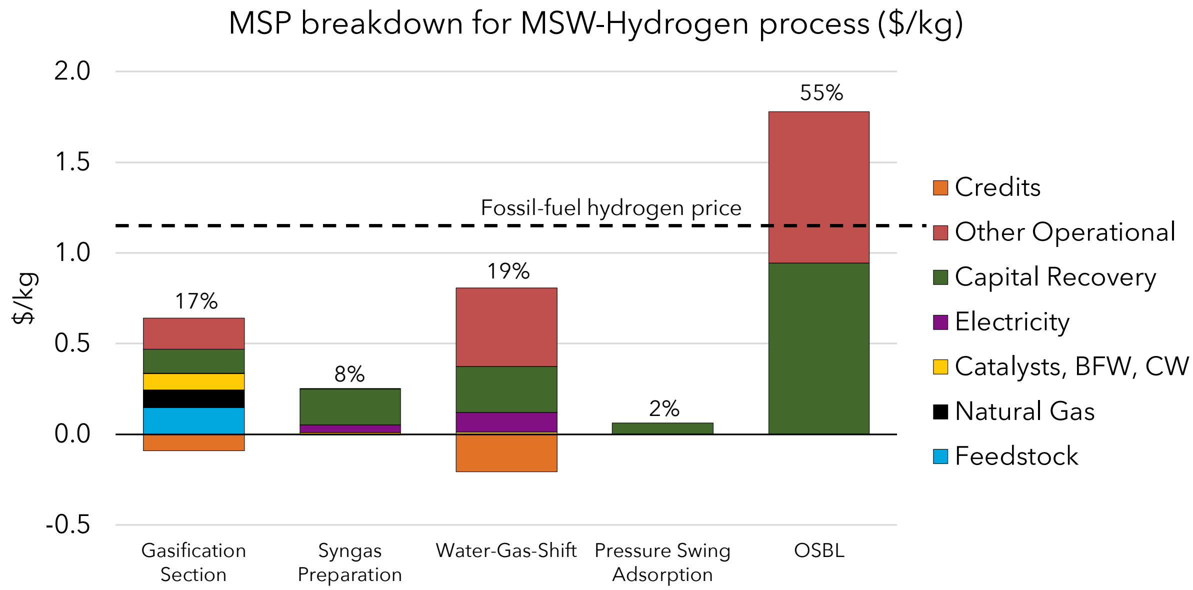

Fig. S7. *Hydrogen MSP breakdown for MSW-Hydrogen process*

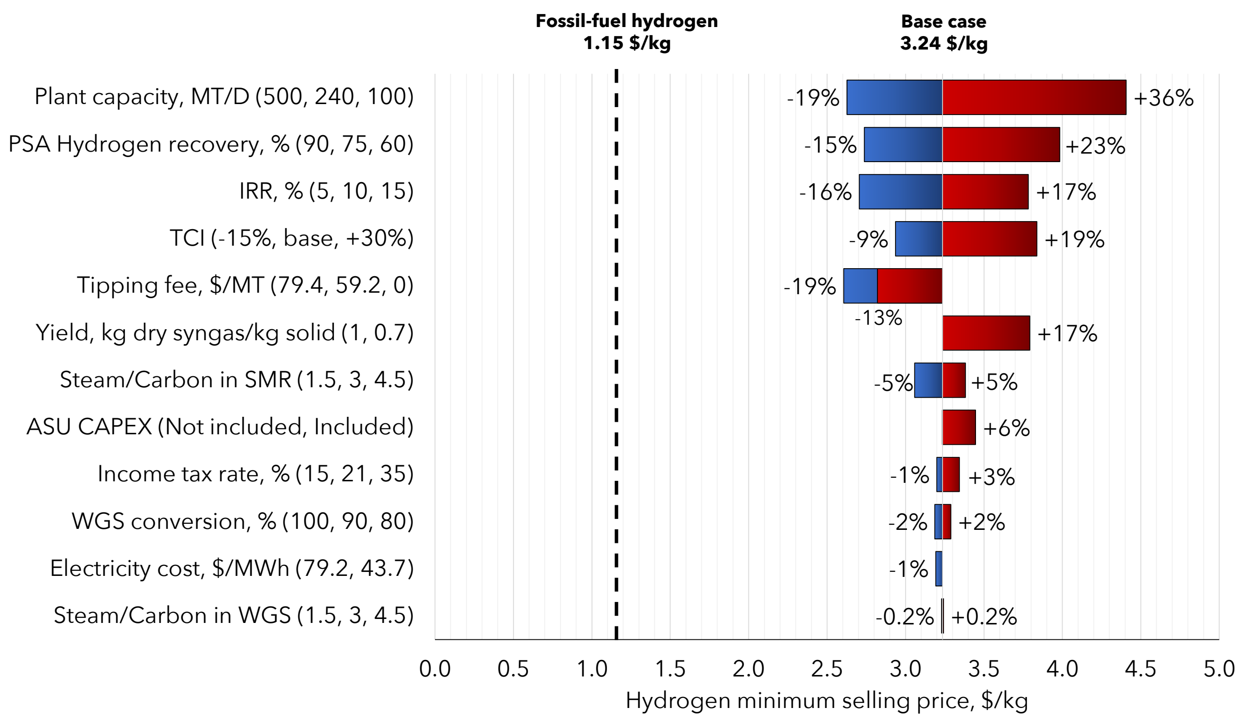

Fig. S8. *Sensitivity analysis for MSW-Hydrogen process.* Plant capacity and PSA Hydrogen recovery efficiency have largest impact on hydrogen MSP.

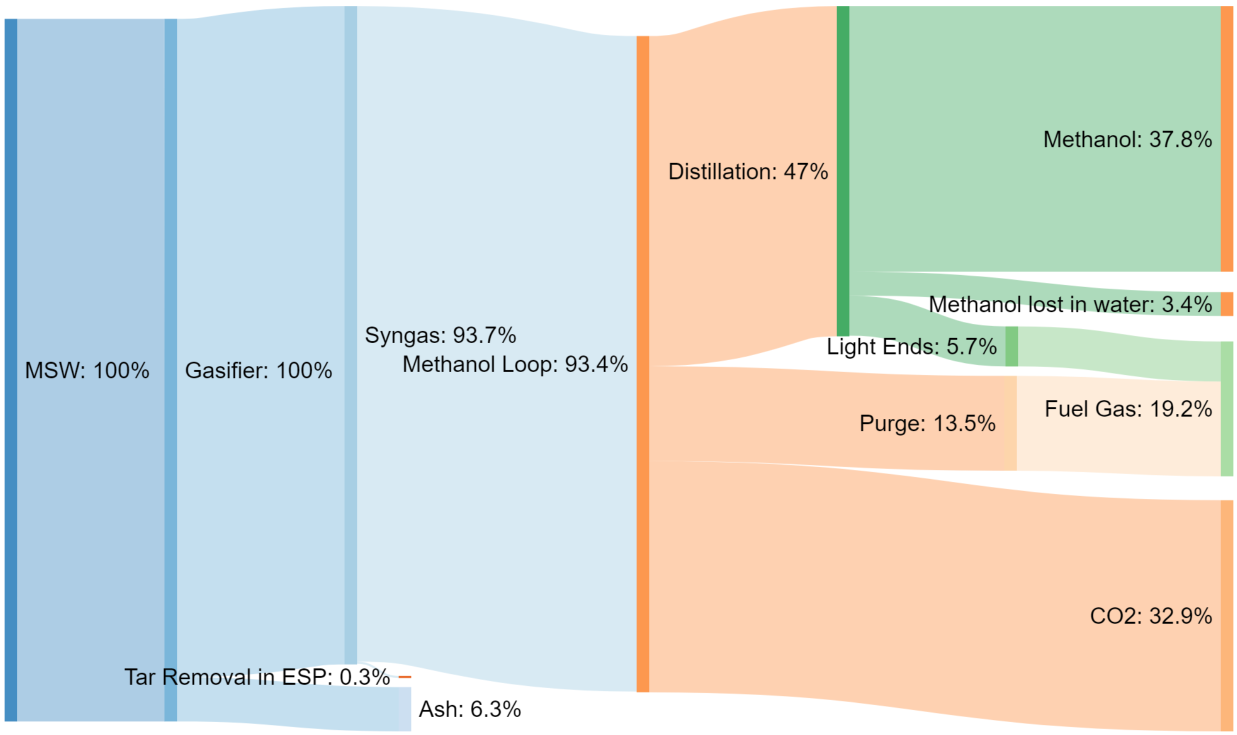

Fig. S9. *Sankey diagram for carbon balance in the MSW-Methanol process.* The carbon efficiency is low (37.8%), mainly due to loss of carbon as CO_2 from the gasifier which is removed in CO_2 removal prior to methanol synthesis.

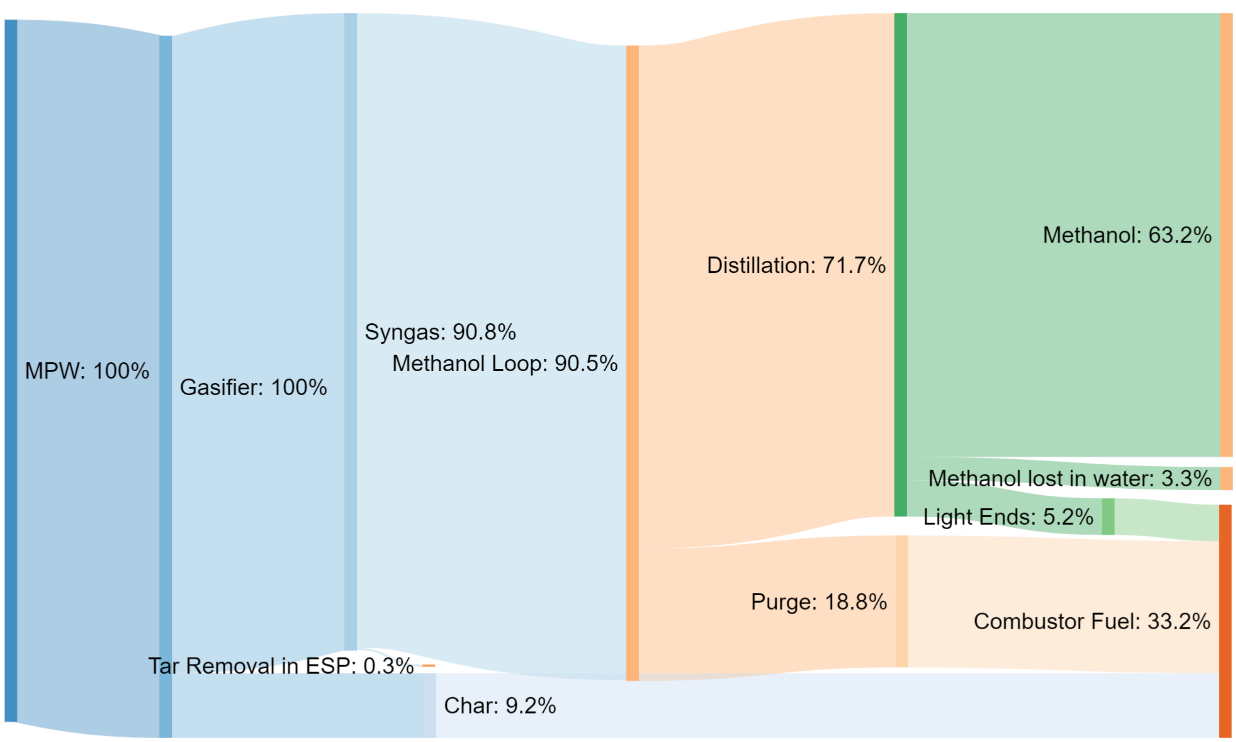

Fig. S10. *Sankey diagram for carbon balance in the MPW-Methanol process.* Carbon efficiency is 63.2%. Main losses are from the purge stream in methanol synthesis loop and the char formed in the gasifier.

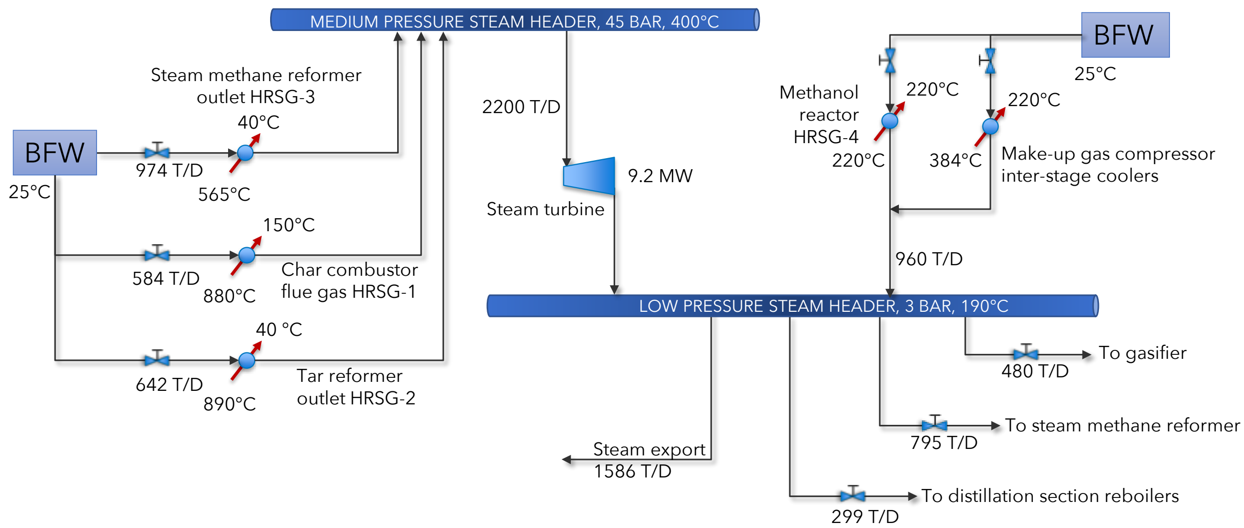

Fig. S11. *Steam system for MPW-Methanol process.* The low-pressure level for steam was chosen since that is the pressure at which steam is needed for the steam methane reformer and the distillation section reboilers. The flowrates of BFW through each exchanger is adjusted to meet the final steam quality requirements. Based on the steam flow shown here, appropriate penalties and credits were taken when performing the GHG emissions analysis by the Materials Flow through Industry (MFI) tool and Life Cycle Analysis (LCA).

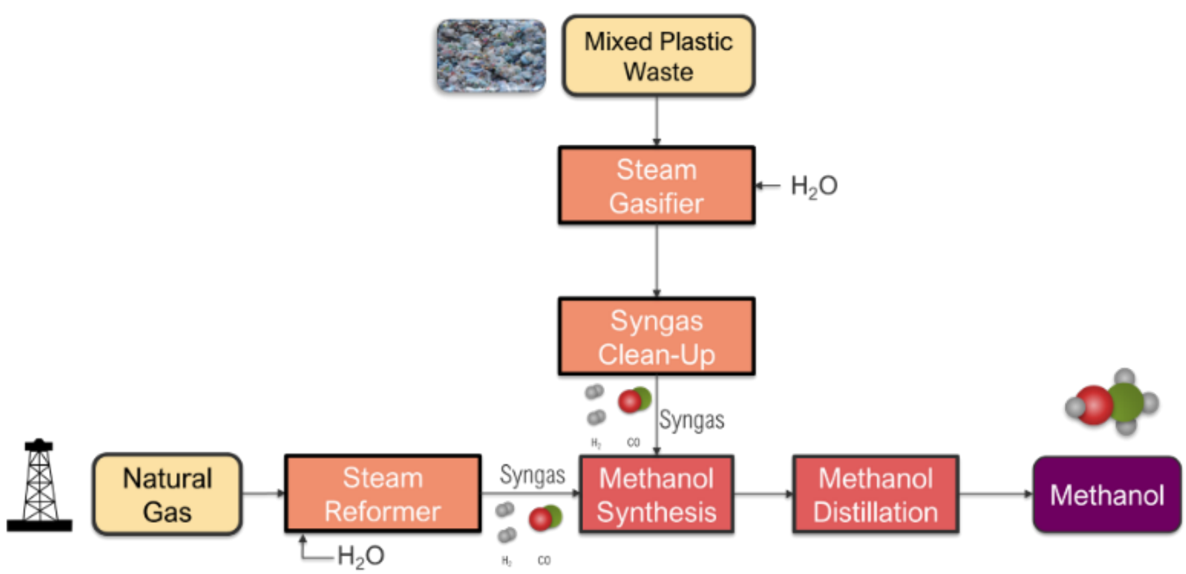

Fig. S12. *Downstream processing of syngas remains the same irrespective of feedstock.*

S2. Supporting Tables

Table S1. *Mass balance around MPW gasifier.* The gasification data for MPW was taken from literature[1] as mentioned in the main text. This table was used to estimate the yield for each gas that was then input into the ASPEN Plus for modeling the gasifier. This applies to the gasifier in MPW-Methanol and MPW-Hydrogen processes.

Syngas yield (dry basis)	**1.38**	**kg/kg**					
	2.1	**Nm^3/kg**					
Gas	**%**	**Nm^3/kg plastic**	**Nm^3/h**	**MW**	**moles/h**	**kg/h**	**Yield for Aspen Plus RYIELD model**
H_2	47	0.987	9,870	2.0	440,616.9	888.2	2.96E-02
CO	22	0.462	4,620	28.0	206,246.2	5,777.0	1.93E-01
CO_2	5	0.105	1,050	44.0	46,874.1	2,062.9	6.88E-02
CH_4	17	0.357	3,570	16.0	159,372.1	2,556.8	8.52E-02
C_2H_4	8	0.168	1,680	28.1	74,998.6	2,104.0	7.01E-02
C_3H_8	1	0.021	210	44.1	9,374.8	413.4	1.38E-02
Water out	14520	kg/h				14,520.0	4.84E-01
C in tar from tar analysis[1] (assumed to be $C_{10}H_8$, naphthalene)	31	g/Nm^3	65.1	g/kg plastic		651.0	2.17E-02
C in Char, combustion from mass balance[1]						944.5	3.15E-02
Ash						10.0	3.33E-04
Ash2						72.3	2.41E-03

The MPW feedstock used in the gasification experiment[1] did not have impurities or contaminants with the PE, PP feed. This is corroborated from **Table S1**, where the ash content after gasification is low (0.8% of the solid feed). Hence, while selecting the feedstock prices, the natural HDPE prices were selected for PE which have much stricter requirements and are more expensive than colored HDPE bales.[2]

Table S2. Operating cost data. The operating costs are based on average of 2015-2019 values from an industry database.

Utility / Operating cost	**Value**	**Units**
Natural gas	150.4	$/MT
Oxygen import	65.78	$/MT
Cyclone and ESP operation[3]	17.8	$/scfm
Olivine cost	275.88	$/MT
Boiler feed water	0.29692	$/MT
Electricity (fossil-fuel)	43.7	$/MWh
Electricity (PV)	79.2	$/MWh
Cooling water (assuming 10 °C ΔT)	3.6509	$/Gcal

Table S3. *CAPEX breakdown by process sections for MPW-Methanol process.* BFW-Boiler feedwater, HRSG-Heat recovery steam generator, SMR-Steam methane reformer.

Equipment Name	Installation Factor	Capital cost (2016$)	
		Equipment Cost	Installed Cost
Gasification			
Circulating fluidized bed reactor system	2.47	1,767,262	4,365,138
Syngas knock-out pot	2.47	45,238	108,376
Shredder	2	157,725	274,408
BFW pump	2.47	109,302	197,161
HRSG-2	2.47	133,318	240,482
HRSG-1	2.47	125,144	225,738
HRSG-3	2.47	174,724	315,172
Balance of plant		251,271	572,798
		2.76M	**6.30M**
Syngas preparation			
Syngas compressor (1-stage)	1.80	2,534,833	4,562,700
ZnO bed	2.47	22,890	56,538
SMR Product/Feed exchanger	2.47	70,360	173,789
SMR feed heater	2.47	27,393	67,660
SMR	2.47	3,730,077	9,213,290
High pressure syngas separator	2.47	64,846	160,170
Balance of plant		645,040	1,423,415
		7.1M	**15.66M**
Methanol synthesis			
Make up gas compressor	1.80	10,635,793	19,144,428
Steam turbine	1.80	5,618,200	10,282,643
Methanol synthesis reactor/HRSG	2.60	387,973	1,008,729
Methanol product/HRSG	2.47	99,432	245,598
Methanol knock-out pot	2.47	66,940	165,342
Recycle gas compressor	1.80	1,277,870	2,300,166
Recycle gas heater/Inter-stage cooler	2.47	37,825	93,427
Inter-stage cooler/HRSG-2	2.47	21,439	52,955
Inter-stage cooler/HRSG-3	2.47	37,603	92,880
BFW pump	2.47	83,145	205,369
Balance of plant		1,826,622	3,359,154
		20.09M	**36.95M**
Methanol distillation			
Crude methanol exchanger/LPS	2.47	10,446	18,842
Distillation column – 1 (C1)	1.30	125,316	162,911
C1 condensor CW Cooler	2.47	22,758	56,212
Distillation column – 2 (C2)	1.27	281,278	357,223
C1 reboiler/LPS exchanger	2.47	28,008	69,181
C2 condensor CW cooler	2.47	95,306	235,406
C2 reboiler/LPS exchanger	2.47	44,934	110,988
C2 feed CW cooler	2.47	72,337	178,674
C1 combined vapor heater/LPS exchanger	2.47	8,961	22,133
Balance of plant		68,935	121,157
		0.76M	**1.33M**

Total installed cost	**30.7M**	**60.2M**
Outside battery limit		15,060,440
Site development and piping		13,177,885
Total indirect costs		53,088,051
Land and working capital		7,218,407
TOTAL CAPITAL INVESTMENT		**148.8M**

Table S4. *OPEX breakdown for MPW-Methanol process.* ESP-Electrostatic precipitator, NG-natural gas, MUG-make up gas, RG-recycle gas, CW-cooling water.

Variable operating costs ($M/y)				**53.9**
Raw material/utility	Value	Units	$M/y (2016)	
Gasification				
MPW	10,000	kg/h	47.3	
External NG for heating	1,051	kg/h	1.25	
NG in tar reformer	7.9	Gcal/h	0.7	
Cyclone and ESP operation	1,415,427	ft^3/h of gas	0.37	
Make-up olivine cost	297	kg/h	0.65	
BFW	91,625	kg/h	0.21	
BFW pump	162.4	kW	0.06	
Tar reformer catalyst cost			0.09	
Tar reformer catalyst replenishment			1.41	
LPS credit (@$5.7/MT)	65,986	kg/h	-2.97	
Wastewater treatment	9,272	kg/h	0.07	
Hammermill operation power input	116	kWh/MT	0.4	
Disposal of solids	82	kg/h	0.04	
			49.58	
Syngas preparation				
Syngas compression	1976.7	kW	0.68	
ZnO bed catalyst	694,457	ft^3/h of gas	0.01	
SMR feed preheater	0.3	Gcal/h	0.03	
SMR furnace duty	17.0	Gcal/h	1.51	
SMR catalyst	2,961,707	sft^3/h of gas	0.15	
BFW credit	25,494	kg/h	-0.06	
			2.33	
Methanol synthesis				
Compression duty for MUG compressor	11870.6	kW	4.09	
Turbine electricity credit	9205.6	kW	-3.17	
Compression duty for RG compressor	839.7	kW	0.29	
BFW	39,999	kg/h	0.09	
BFW pump	3.7	kW	0	
Methanol synthesis catalyst	3,939,105	sft^3/h of gas	0.21	

			1.51	
Methanol distillation				
C1 condensor (CW)	0.4	Gcal/h	0.01	
C2 condensor (CW)	11.4	Gcal/h	0.33	
Cooling duty for C2 feed cooler	6.1	Gcal/h	0.18	
BFW credit	12,500	kg/h	-0.03	
			0.49	
Fixed operating costs ($M/y)				**8.18**
Labor and supervision ($M/y)			**2.94**	
Position	Salary (2016)	# Required	Total Cost (2016)	
Plant Manager	$164,452	1	$164,452	
Plant Engineer	$78,310	1	$78,310	
Maintenance Supr	$63,767	1	$63,767	
Maintenance Tech	$44,749	6	$268,493	
Lab Manager	$62,648	1	$62,648	
Lab Technician	$44,749	1	$44,749	
Shift Supervisor	$53,699	3	$161,096	
Shift Operators	$44,749	12	$536,985	
Yard Employees	$31,324	4	$125,297	
Clerks & Secretaries	$40,274	1	$40,274	
Total Salaries			$1,546,070	
Labor Burden	90.00%	of Total Salaries	$1,391,463	
Overhead ($M/y)			**5.24**	
Maintenance	3.00%	of FCI	$4,247,044	
Property Insur. & Tax	0.70%	of FCI	$990,977	
TOTAL OPERATING COST ($M/y)				**62.08**

Table S5. *MSP breakdown for MPW-Methanol process.*

MSP breakdown ($/kg)								
Process section	Feedstock	Natural gas	Catalysts, BFW, CW	Electricity	Capital Recovery	Other Operational	Credits	Total
Gasification	0.41	0.02	0.02	0.00	0.01	0.00	-0.03	0.43
Syngas preparation	0.00	0.01	0.00	0.01	0.02	0.00	0.00	0.04
Methanol synthesis	0.00	0.00	0.00	0.04	0.04	0.00	-0.03	0.05
Methanol distillation	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01
OSBL	0.00	0.00	0.00	0.00	0.10	0.07	0.00	0.17
Total	0.41	0.03	0.03	0.04	0.17	0.07	-0.05	**0.70**

Table S6. *Reasoning for choice of bounds for sensitivity analysis for MPW-Methanol process.* This table applies to the MPW-Hydrogen process too.

Sensitivity	Reasoning for choice of bounds	MSP-lower end, $/kg	MSP-higher end, $/kg
Yield, kg dry syngas/kg solid (1.4, 1.0)	The base case yield is 1.4 kg dry syngas/kg solid. The lower point of 1.0 kg dry syngas/kg solid represents a reduction in gasifier yield, which could be due to design/operation issues with the gasifier. A yield greater than 1.4 kg dry syngas/kg solid results in a mass balance error for carbon and hence only a lower point was considered. Theoretically, much higher yield (2.3 kg dry syngas/kg MPW) is possible through steam gasification, if all solid MPW was converted only to ($CO+H_2$) without any side-products or losses by char formation. But not all carbon atoms end up as CO in the gasifier, which is the main reason of increase in mass yield. Only about 29% of total carbon input ends in CO.	0.7	0.91
Feedstock cost, $/kg (0.40, 0.60, 0.80)	This range was chosen based on prices in Recycling Markets[2]. Higher prices could be due to higher labor and sorting costs in certain geographic locations.	0.56	0.83
Plant capacity, MT/D (500, 240, 100)	Recent plants utilizing the MPW feedstock are in the 120 – 300 MT/D range and hence plant capacity bounds were chosen accordingly.	0.64	0.8
Methanol loop purge fraction, % (5, 15, 25)	This represents variability in amount of gas purged from the synthesis loop to the fuel gas system. A higher conversion to methanol in reactor will enable operators to lower the purge flow and vice versa.	0.63	0.77
Single pass conversion to methanol, % (50, 40, 30)	This variability is used to span the range of conversions of different catalysts for methanol synthesis.	0.65	0.77
IRR, % (5, 10, 15)	This is a financial parameter.	0.64	0.75
TCI (-15%, base, +30%)	The variability in TCI due to inflation, location and price fluctuations is represented in this sensitivity.	0.67	0.76
Steam export credit (with, without)	This represents a case where the steam export credit is not accounted for, which is a plausible scenario if the plant is located in a remote region without a steam customer in close proximity to the plant.	0.7	0.72
Steam/Carbon in SMR (1.5, 3, 4.5)	The SMR catalyst has Steam/Carbon (S/C) limitations to avoid coke formation. A higher S/C ratio reduces risk of coke formation but increase energy requirement. This variability is captured in the range shown as per industrial conditions[4] (3 – 4.5) and the 1.5 represents an optimistic case.	0.69	0.71
Income tax rate, % (15, 21, 35)	This is a financial parameter.	0.69	0.71
Methanol catalyst cost, $/kg (1.8, 18, 180)	This order-of-magnitude change is used as sensitivity to span the catalyst cost domain for different metal catalysts.	0.69	0.71
Electricity cost, $/MWh (43.7, 79.2)	The base case electricity cost is 43.7 $/MWh. The 79.2 $/MWh point is used as a case for higher utility cost from different renewable sources[5].	0.7	0.71
Methanol conversion to DME, % (5, 10, 15)	This sensitivity represents the case where a methanol catalyst is producing low or higher side-products (simulated as DME here).	0.69	0.7

Table S7. *Mass balance around MSW gasifier.* The gasification data for MPW was taken from literature[6] as mentioned in the main text. This table was used to estimate the yield for each gas that was then input into the ASPEN Plus for modeling the gasifier. This applies to the gasifier in MSW-Methanol and MSW-Hydrogen processes.

Syngas yield (dry basis)	**1.0**	**kg/kg**				
	0.8	**Nm^3/kg**				
Gas	**%**	**Nm^3**	**kmol**	**MW, kg/kmol**	**MT/D**	**Yield for Aspen Plus RYIELD model**
N_2	0.81	1320.776	58.9	28.0	1.7	4.17E-03
H_2	11.86	19355.079	863.5	2.0	1.7	4.40E-03
CO	20.36	33231.382	1482.6	28.0	41.5	1.05E-01
CO_2	41.03	66968.245	2987.8	44.0	131.5	3.32E-01
CH_4	9.93	16208.044	723.1	16.0	11.6	2.93E-02
C_2H_6	10.23	16697.220	744.9	30.1	22.4	5.66E-02
C_3H_8	4.99	8136.634	363.0	44.1	16.0	4.04E-02
$C_{10}H_8$	0.71	1157.717	51.7	128.2	6.6	1.67E-02
NH_3	5.99E-02	97.835	4.4	17.0	0.1	1.88E-04
H_2S	8.19E-03	13.371	0.6	34.1	0.0	5.13E-05
COS	8.19E-03	13.371	0.6	60.1	0.0	9.05E-05
HCl	2.00E-04	0.326	0.0	36.5	0.0	1.34E-06

Table S8. *CAPEX breakdown by process sections for MSW-Methanol process.*

Equipment name	Installation factor	Capital cost (2016$)	
		Equipment cost	Installed cost
Gasification			
Fixed bed gasifier system	2.47	2,004,988	4,952,320
Syngas knock-out pot	2.47	29,404	70,443
BFW pump	2.47	69,603	125,551
HRSG-2	2.47	70,839	127,781
HRSG-1	2.47	63,389	114,343
HRSG-3	2.47	42,867	77,324
Balance of plant		243,881	574,217
		2.53M	**6.04M**
Syngas preparation			
Syngas compressor (1-stage)	1.80	1,360,168	2,448,303
LO-CAT absorption vessel	2.47	127,611	315,200
ZnO bed	2.47	17,964	44,372
SMR Product/Feed exchanger	2.47	43,220	106,754
SMR feed heater	2.47	16,017	39,561
Steam methane reformer	2.47	1,274,681	3,148,462
High pressure syngas separator	2.47	36,227	89,480
Balance of plant		287,589	619,213
		3.16M	**6.81M**

Methanol synthesis			
Make up gas compressor	1.80	4,572,339	8,230,210
Steam turbine	1.80	2,423,695	4,435,939
Methanol synthesis reactor/HRSG	2.60	195,537	508,396
Methanol product/HRSG	2.47	41,238	101,857
Methanol knock-out pot	2.47	33,738	83,332
Recycle gas compressor	1.80	349,324	628,783
Recycle gas heater/Inter-stage cooler	2.47	13,221	32,655
Inter-stage cooler/HRSG-2	2.47	10,202	25,198
Inter-stage cooler/HRSG-3	2.47	19,002	46,934
Inter-stage cooler/HRSG-4B	2.47	11,153	27,547
BFW pump	2.47	58,916	145,522
Acid gas removal system	2.61	632,902	1,651,875
Balance of plant		836,127	1,591,825
		9.20M	**17.51M**
Methanol distillation			
Crude methanol exchanger/LPS	2.47	4,773	8,609
Distillation column – 1 (C1)	1.30	54,851	71,306
C1 condensor CW cooler	2.47	13,345	32,961
Distillation column – 2 (C2)	1.27	120,028	152,436
C1 reboiler/LPS Exchanger	2.47	12,408	30,647
C2 condensor CW cooler	2.47	55,710	137,604
C2 reboiler/LPS exchanger	2.47	21,595	53,340
C2 feed CW cooler	2.47	41,215	101,801
C1 combined vapor heater/LPS exchanger	2.47	3,898	9,628
Balance of plant		32,782	59,833
		0.36M	**0.66M**
Total installed cost		**15.39M**	**31.27M**
Outside battery limit			7,823,993
Site development and piping			6,845,994
Total indirect costs			27,579,576
Land and working capital			3,817,277
TOTAL CAPITAL INVESTMENT			**77.36M**

Table S9. *OPEX breakdown for MSW-Methanol process.*

Variable operating costs ($M/y)				**5.20**
Raw material/utility	Value	Units	$M/y (2016)	
Gasification				
Natural gas usage	4.5	Gcal/h	0.40	
Oxygen import	2,085	kg/h	1.08	
Cyclone and ESP operation	652,546	ft^3/h of gas	0.17	
BFW	23,338	kg/h	0.05	
BFW pump	53.2	kW	0.02	
Tar reformer catalyst cost	2,682,176	ft^3/h of gas	0.03	
Tar reformer catalyst replenishment			0.54	
LPS credit (@$5.7/mt)	22,217	kg/h	-1.00	
Waste water treatment	1,354	kg/h	0.01	
Waste disposal	2,367	kg/h	1.10	
			2.41	
Syngas preparation				
Syngas compression	907.8	kW	0.31	
LO-CAT chemicals	2	kg/h	0.00	

ZnO bed catalyst	317,091	ft^3/h of gas	0.00	
SMR catalyst	929,276	sft^3/h of gas	0.05	
BFW credit	6,480	kg/h	-0.02	
			0.35	
Methanol synthesis				
Compression duty for MUG compressor	4132.2	kW	1.42	
Turbine electricity credit	2769.8	kW	-0.95	
Compression duty for RG compressor	166.0	kW	0.06	
BFW	14,083	kg/h	0.03	
BFW pump	1.7	kW	0.00	
Methanol synthesis catalyst	974,520	sft^3/h of gas	0.05	
CO_2 separation unit	5,041	kg/h	1.68	
			2.29	
Methanol distillation				
C1 column condensor (CW)	0.1	Gcal/h	0.00	
C2 column condensor (CW)	3.4	Gcal/h	0.10	
Cooling duty for C-2 feed cooler	1.7	Gcal/h	0.05	
BFW credit	3,537	kg/h	-0.01	
Waste water treatment	1,413	kg/h	0.01	
			0.15	
Fixed operating costs ($M/y)				**5.66**
Labor and supervision ($M/y)			**2.94**	
Position	Salary (2016)	# Required	Total Cost (2016)	
Plant Manager	$164,452	1	$164,452	
Plant Engineer	$78,310	1	$78,310	
Maintenance Supr	$63,767	1	$63,767	
Maintenance Tech	$44,749	6	$268,493	
Lab Manager	$62,648	1	$62,648	
Lab Technician	$44,749	1	$44,749	
Shift Supervisor	$53,699	3	$161,096	
Shift Operators	$44,749	12	$536,985	
Yard Employees	$31,324	4	$125,297	
Clerks & Secretaries	$40,274	1	$40,274	
Total Salaries			$1,546,070	
Labor Burden	90.00%	of Total Salaries	$1,391,463	
Overhead ($M/y)			**2.72**	
Maintenance	3.00%	of FCI	$2,206,366	
Property Insur. & Tax	0.70%	of FCI	$514,819	
TOTAL OPERATING COST ($M/y)				**10.86**

Table S10. *MSP breakdown for MSW-Methanol process.*

MSP breakdown ($/kg)								
Process area	Feedstock	Natural gas	Catalysts, BFW, CW	Electricity	Capital Recovery	Other Operational	Credits	Total
Gasification	0.03	0.01	0.02	0.00	0.02	0.03	-0.03	0.09
Syngas preparation	0.00	0.00	0.00	0.01	0.02	0.00	0.00	0.03
Methanol synthesis	0.00	0.00	0.00	0.04	0.06	0.04	-0.03	0.12
Methanol distillation	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01
OSBL	0.00	0.00	0.00	0.00	0.16	0.15	0.00	0.31
Total	0.03	0.01	0.02	0.05	0.26	0.23	-0.05	**0.55**

Table S11. *Reasoning for choice of bounds for sensitivity analysis for MSW-Methanol process.* This table applies to the MSW-Hydrogen process too.

Sensitivity	Reasoning for choice of bounds	MSP-lower end, $/kg	MSP-higher end. $/kg
Plant capacity, MT/D (500, 240, 100)	Recent plants utilizing MSW feedstock are in the 120 – 300 MT/D range and hence plant capacity bounds were chosen accordingly.	0.45	0.76
Steam/Carbon in SMR, (1, 1.3, 3)	The SMR catalyst has Steam/Carbon (S/C) limitations to avoid coke formation. A higher S/C ratio reduces risk of coke formation but increase energy requirement. This variability is captured in the range shown as per industrial conditions[4] (3 – 4.5) and the 1.5 represents an optimistic case.	0.51	0.73
IRR, % (5, 10, 15)	This is a financial parameter.	0.46	0.64
TCI (-15%, base, +30%)	The variability in TCI due to inflation, location and price fluctuations is represented in this sensitivity.	0.50	0.65
Tipping fee, $/MT (79.4, 59.2, 0)	The base case assumes the MSW feed in RDF form is available at zero cost. The US average landfilling fee is $59.2/MT and the Pacific region landfilling fee is $79.4/MT.[7] A feedstock preparation cost[8] of $20/MT was used for the cases where the tipping fee was considered.	0.43	0.47
Methanol loop purge fraction, % (5, 15, 25)	This represents variability in amount of gas purged from the synthesis loop to the fuel gas system. A higher conversion to methanol in reactor will enable operators to lower the purge flow and vice versa.	0.50	0.60
Yield, kg dry syngas/kg solid (1, 0.7)	The base case yield is 1.0 kg dry syngas/kg solid. The lower point of 0.7 kg dry syngas/kg solid represents a reduction in gasifier yield, which could be due to design/operation issues with the gasifier.	0.55	0.65
Single pass conversion to methanol, % (50, 40, 30)	This variability is used to span the range of conversions of different catalysts for methanol synthesis.	0.52	0.61
Methanol conversion to DME, % (5, 10, 15)	This sensitivity represents the case where a methanol catalyst is producing low or higher side-products (simulated as DME here).	0.53	0.57

ASU CAPEX (Not included, Included)	This sensitivity variable was added to study the effect of including the Air separation unit (ASU) CAPEX in the capital costs.	0.55	0.60
Income tax rate, % (15, 21, 35)	This is a financial parameter.	0.55	0.57
Electricity cost, $/MWh (43.7, 79.2)	The base case electricity cost is 43.7 $/MWh. The 79.2 $/MWh point is used as a case for higher utility cost from different renewable sources.[5]	0.55	0.57

Table S12. *CAPEX breakdown by process sections for MPW-Hydrogen process.*

Equipment name	Installation factor	Capital cost (2016$)	
		Equipment cost	Installed cost
Gasification			
Circulating fluidized bed reactor system	2.47	1,767,262	4,365,138
Syngas knock-out pot	2.47	45,238	108,376
Hammermill/Shredder	1.7	144,154	245,061
BFW pump	2.47	113,552	204,828
HRSG-2	2.47	133,318	240,482
HRSG-1	2.47	160,162	288,905
HRSG-3	2.47	174,724	315,172
Balance of plant		255,198	579,882
		2.79M	**6.35M**
Syngas preparation			
Syngas compressor (1-stage)	1.8	2,534,833	4,562,700
ZnO bed	2.47	22,890	56,538
SMR Product/Feed exchanger	2.47	70,360	173,789
SMR feed heater	2.47	27,393	67,660
Steam methane reformer	2.47	3,820,269	9,436,066
High pressure syngas separator	2.47	64,846	160,170
Balance of plant		654,059	1,445,692
		7.19M	**15.90M**
Water-gas-shift			
Main compressor	1.8	6,233,501	11,220,302
Steam turbine	1.8	6,846,431	12,530,599
BFW pump	2.47	78,822	194,690
Water-gas-shift reactor	2.47	43,273	106,885
WGS HRSG	2.47	154,357	381,261
Cooling water exchanger (for WGS reaction mixture)	2.47	68,165	122,957
Knock-out pot	2.47	70,941	175,225
Balance of plant		1,349,549	2,473,192
		14.85M	**27.21M**
Pressure swing adsorption			
Pressure swing adsorption unit	1.9	4,427,976	8,413,154
Balance of plant		442,798	841,315
		4.87M	**9.25M**
	Total	**29.72M**	**58.74M**
Outside battery limit			14,685,223
Site development and piping			12,849,570
Total indirect costs			51,765,411

Land and working capital	7,042,055
TOTAL CAPITAL INVESTMENT	**145.08M**

Table S13. *OPEX breakdown for MPW-Hydrogen process.*

Variable operating costs ($M/y)				**51.76**
Raw material/utility	Value	Units	$M/y (2016)	
Gasification				
MPW	10,000	kg/h	47.30	
External NG for heating	1,866	kg/h	2.21	
NG in tar reformer	7.9	Gcal/h	0.70	
Cyclone and ESP operation	1,415,427	ft^3/h of gas	0.37	
Make-up olivine cost	297	kg/h	0.65	
BFW	102,854	kg/h	0.24	
BFW pump	179.5	kW	0.06	
Tar reformer catalyst cost	7,017,113	ft^3/h of gas	0.09	
Tar reformer catalyst replenishment			1.41	
LPS credit (@$5.7/MT)	49,717	kg/h	-2.23	
Wastewater treatment	9,272	kg/h	0.07	
Hammermill operation power input	116	kWh/MT	0.40	
Disposal of solids	82	kg/h	0.04	
			51.31	
Syngas preparation				
Syngas compression	1976.7	kW	0.68	
ZnO bed catalyst	694,457	ft^3/h of gas	0.01	
SMR feed preheater	0.3	Gcal/h	0.03	
SMR furnace duty	17.6	Gcal/h	1.57	
SMR catalyst	2,961,707	sft^3/h of gas	0.15	
BFW credit	25,494	kg/h	-0.06	
			2.38	
Water-gas-shift				
Compression duty for main compressor	6087.3	kW	2.10	
Turbine electricity credit	12210.1	kW	-4.21	
BFW	34,023	kg/h	0.08	
BFW pump	30.8	kW	0.01	
Water-gas-shift catalyst	55,608	sft^3/h of gas	0.00	
Raw hydrogen gas cooler	5.3	Gcal/h	0.15	
BFW credit	29,559	kg/h	-0.07	
			-1.93	
Fixed operating cost ($M/y)				**8.05**
Labor and supervision ($M/y)			**2.94**	
Position	Salary (2016)	# Required	Total Cost (2016)	
Plant Manager	$164,452	1	$164,452	
Plant Engineer	$78,310	1	$78,310	
Maintenance Supr	$63,767	1	$63,767	
Maintenance Tech	$44,749	6	$268,493	
Lab Manager	$62,648	1	$62,648	
Lab Technician	$44,749	1	$44,749	
Shift Supervisor	$53,699	3	$161,096	
Shift Operators	$44,749	12	$536,985	
Yard Employees	$31,324	4	$125,297	
Clerks & Secretaries	$40,274	1	$40,274	

Total Salaries			$1,546,070	
Labor Burden	90.0%	of Total Salaries		
Overhead ($M/y)			**5.11**	
Maintenance	3.0%	of FCI	$4,141,233	
Property Insur. & Tax	0.7%	of FCI	$966,288	
TOTAL OPERATING COST ($M/y)				**59.8**

Table S14. *MSP breakdown for MPW-Hydrogen process.*

MSP Breakdown ($/kg)								
Process Area	Feedstock	Natural gas	Catalysts, BFW, CW	Electricity	Capital Recovery	Other Operational	Credits	Total
Gasification	2.06	0.13	0.10	0.00	0.07	0.02	-0.10	2.29
Syngas preparation	0.00	0.07	0.01	0.03	0.18	0.00	0.00	0.28
Water-gas-shift	0.00	0.00	0.01	0.09	0.30	0.00	-0.19	0.22
Pressure swing adsorption	0.00	0.00	0.00	0.00	0.10	0.00	0.00	0.10
OSBL	0.00	0.00	0.00	0.00	0.16	0.35	0.00	0.51
Total	2.06	0.20	0.12	0.12	0.82	0.37	-0.29	**3.41**

Table S15. *CAPEX breakdown by process sections for MSW-Hydrogen process.*

Equipment name	Installation factor	Capital cost (2016$)	
		Equipment cost	Installed cost
Gasification			
Fixed bed gasifier system	2.47	2,437,639	6,020,968
Syngas knock-out pot	2.47	29,404	70,443
BFW pump	2.47	81,250	146,562
HRSG-2	2.47	120,181	216,786
HRSG-1	2.47	63,389	114,343
HRSG-3	2.47	50,079	90,334
Balance of plant		293,967	693,384
		3.08M	**7.35M**
Syngas preparation			
Syngas compressor (1-stage)	1.80	1,360,168	2,448,303
LO-CAT system	2.61	1,082,436	2,825,159
ZnO bed	2.47	17,964	44,372
SMR Product/Feed exchanger	2.47	43,220	106,754
SMR feed heater	2.47	16,017	39,561
Steam methane reformer	2.47	1,947,472	4,810,255
High pressure syngas separator	2.47	46,691	115,326
Balance of plant		451,397	1,038,973
		4.97M	**11.43M**
Water-gas-shift			
Main compressor	1.8	2,834,488	5,102,078
CO_2 removal system	2.61	673,246	1,757,172
Steam turbine	1.8	3,365,844	6,160,296

BFW pump	2.47	45,231	111,720
Water-gas-shift reactor	2.47	16,770	41,421
WGS HRSG	2.47	52,384	129,389
Cooling water exchanger (for WGS reaction mixture)	2.47	37,726	68,052
Knock-Out Pot	2.47	25,692	63,459
Balance of plant		705,138	1,343,359
		7.76M	**14.78M**
Pressure swing adsorption			
Pressure swing adsorption unit	1.90	1,720,678	3,269,288
Balance of plant		172,068	326,929
		1.89M	**3.60M**
Outside battery limit			9,357,273
Site development and piping			8,187,614
Total indirect costs			32,984,388
Land and working capital			4,537,918
TOTAL CAPITAL INVESTMENT			**92.5M**

Table S16. *OPEX breakdown for MSW-Hydrogen process.*

Variable operating costs ($M/y)				**6.04**
Raw material/utility	Value	Units	$M/y (2016)	
Gasification				
Oxygen	2,085	kg/h	1.08	
Natural gas usage	7.9	Gcal/h	0.73	
Cyclone and ESP operation	652,546	ft^3/h of gas	0.17	
BFW	37,300	kg/h	0.09	
BFW pump	76.8	kW	0.03	
Tar reformer catalyst cost	2,682,176	ft^3/h of gas	0.03	
Tar reformer catalyst replenishment			0.54	
LPS credit (@$5.7/mt)	15,098	kg/h	-0.68	
Wastewater treatment	1,354	kg/h	0.01	
			2.00	
Syngas preparation				
Syngas compression	907.8	kW	0.31	
LO-CAT chemicals	2	kg/h	0.00	
ZnO bed catalyst	317,091	ft^3/h of gas	0.00	
SMR catalyst	1,394,272	sft^3/h of gas	0.07	
BFW credit	15852	kg/h	-0.04	
			0.35	
Water-gas-shift				
Compression duty for main compressor	2273.0	kW	0.78	
Turbine electricity credit	4427.8	kW	-1.53	
BFW	6,321	kg/h	0.01	
BFW Pump	9.6	kW	0.00	
Water gas shift catalyst	800,565	sft^3/h of gas	0.04	
Raw hydrogen gas cooler	1.4	Gcal/h	0.04	
BFW credit	4,710	kg/h	-0.01	
CO_2 separation	9,729	kg/h	3.23	
			2.58	
Fixed operating cost ($M/y)				**6.19**
Labor and supervision ($M/y)			**2.94**	

Position	Salary (2016)	# Required	Total Cost (2016)	
Plant Manager	$164,452	1	$164,452	
Plant Engineer	$78,310	1	$78,310	
Maintenance Supr	$63,767	1	$63,767	
Maintenance Tech	$44,749	6	$268,493	
Lab Manager	$62,648	1	$62,648	
Lab Technician	$44,749	1	$44,749	
Shift Supervisor	$53,699	3	$161,096	
Shift Operators	$44,749	12	$536,985	
Yard Employees	$31,324	4	$125,297	
Clerks & Secretaries	$40,274	1	$40,274	
Total Salaries			$1,546,070	
Labor Burden	90.0%	of Total Salaries		
Overhead ($M/y)			**3.25**	
Maintenance	3.0%	of FCI	$2,638,751	
Property Insur. & Tax	0.7%	of FCI	$615,709	
TOTAL OPERATING COST ($M/y)				**12.2**

Table S17. *MSP breakdown for MSW-Hydrogen process.*

MSP Breakdown ($/kg)								
Process area	Feedstock	Natural gas	Catalysts, BFW, CW	Electricity	Capital Recovery	Other Operational	Credits	Total
Gasification	0.15	0.10	0.09	0.00	0.13	0.17	-0.09	0.55
Syngas preparation	0.00	0.00	0.01	0.04	0.20	0.00	0.00	0.24
Water-gas-shift	0.00	0.00	0.01	0.11	0.25	0.44	-0.21	0.60
Pressure swing adsorption	0.00	0.00	0.00	0.00	0.06	0.00	0.00	0.06
OSBL	0.00	0.00	0.00	0.00	0.95	0.83	0.00	1.78
Total	0.15	0.10	0.11	0.15	1.59	1.44	-0.30	**3.24**

Table S18. *Material Flows through Industry (MFI) analysis results for MPW-Methanol and MSW-Methanol cases.*

METHANOL					
	Fossil-fuel methanol	MPW	MSW	MSW (with credit for avoiding landfill)	Unit
TOTAL SUPPLY CHAIN ENERGY	**37.0**	**17.8**	**9.9**		MJ/kg methanol
Process Fuel	6.1	12.3	3.7		MJ/kg methanol
Fuel for Electricity	0.3	4.3	5.2		MJ/kg methanol
Renewable Electricity	0.0	0.3	0.4		MJ/kg methanol
Fuel for Transportation	0.7	0.4	0.5		MJ/kg methanol
Fuel as Chemical Feedstocks	29.8	0.5	0.1		MJ/kg methanol
GHG - Process Fuel	0.3	0.8	1.3	1.3	kg CO_{2e}/kg methanol
GHG - Electricity Generation	0.0	0.3	0.4	0.4	kg CO_{2e}/kg methanol
GHG - Transportation	0.1	0.0	0.0	0.0	kg CO_{2e}/kg methanol
GHG - After applying landfill credit				-2.6	kg CO_{2e}/kg methanol
TOTAL GHG	**0.4**	**1.1**	**1.7**	**-0.9**	kg CO_{2e}/kg methanol

Table S19. *Material Flows through Industry (MFI) analysis results for MPW-Hydrogen and MSW-Hydrogen cases.*

HYDROGEN					
	Fossil-fuel hydrogen	MPW	MSW	MSW (with credit for avoiding landfill)	Unit
TOTAL SUPPLY CHAIN ENERGY	**174.3**	**75.8**	**31.2**		MJ/kg hydrogen
Process Fuel	156.5	76.6	32.7		MJ/kg hydrogen
Fuel for Electricity	4.1	-4.1	-3.6		MJ/kg hydrogen
Renewable Electricity	0.3	-0.2	-0.3		MJ/kg hydrogen
Fuel for Transportation	3.2	1.5	2.3		MJ/kg hydrogen
Fuel as Chemical Feedstocks	10.2	2.0	0.0		MJ/kg hydrogen
GHG - Process Fuel	8.8	12.9	15.7	15.7	kg CO_{2e}/kg hydrogen
GHG - Electricity Generation	0.3	-0.4	-0.3	-0.3	kg CO_{2e}/kg hydrogen
GHG - Transportation	0.3	0.1	0.2	0.2	kg CO_{2e}/kg hydrogen
GHG - After applying landfill credit				-13.1	kg CO_{2e}/kg hydrogen
TOTAL GHG	**9.4**	**12.8**	**15.6**	**2.6**	kg CO_{2e}/kg hydrogen

Table S20. *LCA results for MPW-Methanol and MPW-Hydrogen processes compared to their fossil-fuel counterparts.*

	Impact category										
	Acidification kg SO_2 eq/kg	**Carcinogenics** CTUh/kg	**Eco-toxicity** CTUe/kg	**Eutrophication** kg N eq/kg	**Fossil fuel depletion** MJ surplus/kg	**Global warming** kg CO_2 eq./kg	**Non carcinogenic** CTUh/kg	**Ozone depletion** kg CFC-11/kg	**Respiratory effects** kg PM2.5 eq/kg	**Smog** kg O_3 eq/kg	**Water use** m^3/kg
MPW-methanol											
TOTAL	**0.0017**	**3.13E-8**	**6.66**	**0.0032**	**0.85**	**0.736**	**1.52E−7**	**1.34E−7**	**0.0010**	**−0.0058**	**5.24**
Standard deviation	***0.0008***	***1.86E-8***	***3.29***	***0.0018***	***0.441***	***0.118***	***1.14E−7***	***7.60E−8***	***0.0002***	***0.0051***	***3.73***
Process emissions	0	0	0.138	0	0	1.07	5.5E−10	0	2.68E−8	6.8E−5	0
Cooling water	3.96E−6	2.04E−10	0.015	3.36E−6	0.0009	0.0009	4.3E−10	8.08E−11	1.21E−6	5.7E−5	4.98
Boiler feed water	2.08E−7	1.07E−11	7.82E−4	1.76E−7	4.97E−5	4.94E−5	2.3E−11	4.23E−12	6.33E−8	3.0E−6	0.26
Natural gas	0.0033	4.66E−9	0.614	0.0002	1.915	0.0984	2.34E−8	1.75E−7	0.0003	0.0055	0.007
MPW feedstock	0.00037	6.13E−9	3.50	0.0012	0.2613	0.0710	6.68E−8	2.99E−8	0.00014	0.0043	0.016
Catalysts	9.63E−5	4.58E−9	0.588	0.0001	0.0057	0.0239	2.53E−8	6.08E−10	3.05E−5	0.0009	0.001
Infrastructure	3.83E−8	1.28E−12	0.0003	5.34E−8	3.26E−6	4.34E−6	1.1E−11	3.11E−13	9.06E−9	2.93E−7	1.2E−6
Electricity	0.0011	2.37E−8	2.70	0.0024	0.0230	0.302	8.08E−8	3.21E−8	0.0009	0.0077	0.050
Steam (co-product)	−0.003	−1.2E−8	−1.14	−0.008	−1.56	−0.832	−5.0E−8	−1.0E−7	−0.0003	−0.024	−0.044
Ash disposal	6.72E−7	4.00E−9	0.216	1.52E−5	0.0002	0.0002	1.55E−9	1.62E−11	1.06E−7	1.20E−5	3.5E−5
Wastewater	4.91E−6	1.68E−10	0.0127	3.11E−5	0.0005	0.0006	2.99E−9	4.21E−11	9.39E−7	4.83E−5	−0.04
MPW-hydrogen											
TOTAL	**0.0091**	**9.70E-9**	**15.6**	**−2.51E-5**	**7.18**	**10.8**	**2.61E−7**	**8.16E−7**	**−0.00076**	**−0.047**	**8.42**
Standard deviation	***0.0049***	***8.46E-9***	***10.5***	***0.0004***	***2.34***	***0.456***	***2.96E−7***	***4.93E−7***	***0.0007***	***0.0202***	***8.31***
Process emissions	0	0	0	0	0	13.5	0	0	1.36E−7	0.0003	0
Cooling water	5.98E−6	3.09E−10	0.0226	5.08E−6	0.0014	0.0014	6.5E−10	1.22E−10	1.82E−6	8.61E−5	7.53
Boiler feed water	9.18E−7	4.74E−11	0.0035	7.79E−7	0.0002	0.0002	1.0E−10	1.87E−11	2.80E−7	1.32E−5	1.16
Natural gas	0.0212	2.99E−8	3.94	0.0010	12.26	0.630	1.50E−7	1.12E−6	0.0016	0.0353	0.042
MPW feedstock	0.00188	3.11E−8	17.7	0.0006	1.32	0.360	3.39E−7	1.51E−7	0.0007	0.0220	0.080
Catalysts	0.00047	2.26E−8	2.65	0.0005	0.028	0.121	1.13E−7	3.05E−9	0.0002	0.0042	0.007
Infrastructure	1.95E−7	6.51E−12	0.0013	2.72E−7	1.66E−5	2.21E−5	5.8E−11	1.58E−12	4.62E−8	1.49E−6	6.0E−6
Steam (co-product)	−0.0122	−4.6E−8	−4.33	−0.0029	−5.96	−3.17	−1.9E−7	−4.0E−7	−0.001	−0.093	−0.166
Electricity (co-product)	−0.0023	−4.9E−8	−5.58	−0.0051	−0.475	−0.634	−1.7E−7	−6.6E−8	−0.002	−0.016	−0.104
Ash disposal	3.40E−6	2.03E−8	1.09	7.71E−5	0.0008	0.0013	7.85E−9	8.20E−11	5.37E−7	6.10E−5	0.0002
Wastewater	1.58E−5	5.39E−10	0.041	9.98E−5	0.0016	0.0018	9.58E−9	1.35E−10	3.01E−6	0.0002	−0.126
Fossil-based methanol											
TOTAL	**0.0057**	**1.98E−8**	**3.94**	**0.00117**	**4.81**	**0.576**	**1.56E−7**	**2.16E−7**	**0.000572**	**0.0301**	**0.19**
Standard deviation	***0.0017***	***1.78E-8***	***3.06***	***0.00064***	***0.909***	***0.078***	***1.74E-7***	***9.56E−8***	***0.000125***	***0.0045***	***1.28***
Fossil-based hydrogen											
TOTAL	**0.00693**	**6.32E-8**	**6.68**	**0.00299**	**10.4**	**9.61**	**2.41E−7**	**3.61E−7**	**0.00163**	**0.0867**	**2.0**
Standard deviation	***0.00045***	***4.67E−8***	***4.55***	***0.00143***	***0.053***	***0.285***	***2.05E−7***	***9.18E−8***	***0.00018***	***0.0039***	***6.7***

Table S21. *Heat duties of all major heat transfer equipment.* Refer to **Fig. S13-S26** for respective PFDs.

Tag	Heat duty, Gcal/h	Tag	Heat duty, Gcal/h
Q1	29.18	Q19	-3.62
Q2	18.59	Q20	-0.13
Q3	-7.86	Q21	-3.66
Q4	20.50	Q22	7.75
Q5	-7.67	Q23	9.36
Q6	-0.28	Q24	1.04
Q7	-16.99	Q25	-0.12
Q8	31.07	Q26	-0.53
Q9	27.48	Q27	0.13
Q10	-0.41	Q28	-0.09
Q11	-1.86	Q29	1.70
Q12	0.44	Q30	-1.25
Q13	-0.32	Q31	3.37
Q14	1.70	Q32	25.68
Q15	-3.85	Q33	5.34
Q16	11.43	Q34	4.87
Q17	-6.07	Q35	1.39
Q18	6.53		

S3. MPW-Methanol process

When mixed plastic waste and municipal solid waste gasification pathways are to be compared with commercial benchmarks, the difference in the process lies only in the initial upstream part of the process, up to the syngas generation step as shown in the example of a methanol process in **Fig. S12**. Once the syngas is produced in the desired H_2/CO ratio (2 for methanol) and the catalyst poisons and particulates are removed, the downstream processing remains the same for all processes irrespective of the gasification feedstock. However, syngas is not a traded commodity and hence a market price of syngas for different H_2/CO ratios is not available. Some studies have estimated a syngas price[9,10] but they are only applicable to the respective studies to compare the performance of different pathways. Hence, for a fair comparison, the processes have been modeled until the final production of methanol and hydrogen, and the market price of the final products[11] is compared with the minimum selling price (MSP) of the MPW and MSW-based processes.

The conventional methanol production process from natural gas[12] has been modified to handle an unconventional feedstock like MPW. A simple steam reformer in NG-based plants is replaced by a gasification unit with a combustion reactor that can handle solid feedstock.

Gasification & gas clean-up. The first section in the MPW-Methanol process involves gasification and gas-clean-up and the process flow diagram (PFD) is shown in **Fig. S13**. The indirect gasification design is suitable here since the steam gasification of waste plastics is endothermic, and the circulating olivine provides the energy to drive the gasification reactions. Four streams are combusted in the combustion reactor to supply energy to the gasification reactor, namely (1) char from the gasification reactor, (2) purge stream from the methanol synthesis recycle loop which contains H_2, CO, and methanol, (3) top vapor from the light ends distillation column which contains methanol, H_2, and dimethyl ether, and (4) importing natural gas. The gasification reactor temperature of 834 °C was maintained by adjusting the NG import flow rate. Around 48% of the energy was supplied by importing natural gas in the base case. The hot olivine from the combustion reactor provides the heat to the gasification reactor and is then circulated back to the combustion reactor by solid handling equipment. This design is similar to a continuous catalyst regenerator (CCR) in a refinery[13].

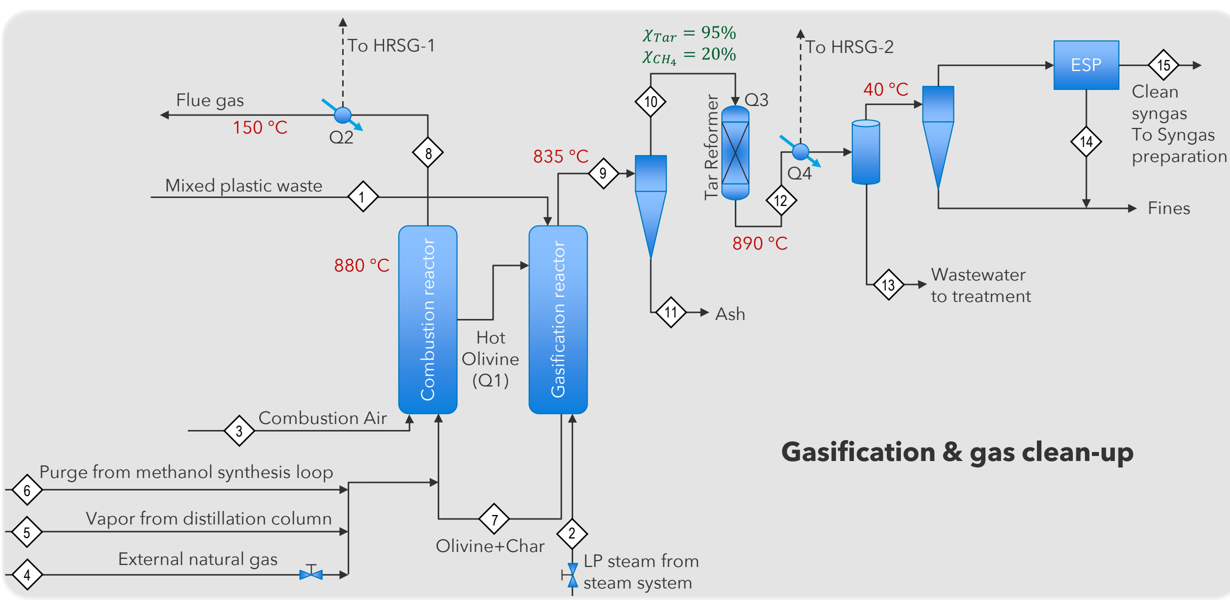

Fig. S13. *Gasification and gas clean-up PFD for MPW-Methanol process*. Refer to Table S12 for heat transfer duties.

The syngas yield is a function of the solid feedstock, gasification agent (steam/oxygen), and syngas composition. The MPW syngas yields are more than MSW syngas yields, with all other parameters remaining the same since the volatile fraction is higher in MPW. Syngas yield is an important parameter that has a strong influence on process economics for MPW.

Based on the experimental results of Wilk and Hofbauer[1], the carbon in the mixed plastic waste ends up in different phases: 82% in the gas phase as CO, CO_2, CH_4, C_2H_4, C_3H_8, 5% in tar[1], 12% in char, and 0.4% in ash. As expected for steam gasification[14], the H_2/CO ratio is high at about 2.14 and is close to the required stoichiometric ratio required for methanol synthesis of 2.

The raw syngas is passed through a cyclone to remove particulates and is then fed into a tar reformer. The tar reformer converts large fused aromatic hydrocarbons like naphthalene to H_2 and CO thereby enhancing syngas yield. Naphthalene ($C_{10}H_8$) was used as the model compound for tar[1]. However, the tar reformer catalyst is designed to maximize tar conversions, and hence the conversion of light hydrocarbon gases such as methane is low. Therefore, an additional steam reformer is needed further downstream to convert light gases to syngas.

In the current model, even after tar reforming, the syngas stream has a tar concentration of 938 mg/Nm^3. The allowable tar content for Fischer-Tropsch synthesis is <1 mg/Nm^3 [15]. This value can be used as a benchmark for other catalytic synthesis pathways. Hence, even after tar reforming, the amount of tar remaining in the gas stream must be reduced. Thus, the cyclone and electrostatic precipitator (ESP) combination is used after the tar reformer for particulate control.

The process is well integrated in terms of heat and material balance. Though a pinch analysis was not carried out, all the major pinch analysis guidelines were followed in the design of the heat exchanger network (HEN). A temperature approach of 15 °C was used in the design of the HEN. As the temperature of the gasifier and steam reformer is in the range of 750-850 °C, and the methanol process make-up gas compressor has a large electricity demand, there is an opportunity for co-generation to reduce the electricity demand as well as satisfy steam demand. **Fig. S11** shows the two-tier steam system used in the MPW-Methanol process. A similar design has been used in all the processes studied with small variations.

The steam system has two headers operating at different pressures. Medium pressure steam (400 °C, 45 bar) is produced in the Heat Recovery Steam Generators (HRSGs) by heat coming from Tar Reformer Outlet, Steam Reformer Outlet, and Char Combustor Flue-gas. The flow rates of BFW are adjusted so the final steam has the same pressure and temperature of 45 bar and 400 °C respectively. All the steam is mixed and runs a turbine to generate electricity. The outlet pressure of the Turbine is fixed at 3 bar, the pressure that is required for the steam reformer. The same Low Pressure (LP) steam is used in the steam reformer as well as reboilers in the distillation columns.

Modeling the gasifier. The simulation step involves base case simulation in ASPEN Plus for the selected process pathways. Specifically for the gasification reactor, non-conventional components have to be used to characterize the solid feeds, Mixed Plastic Waste, and Municipal Solid Waste. Hence the methodology followed is described here for the Mixed Plastic Waste case. The input data is in the form of Proximate and Ultimate Analysis. The Ultimate Analysis input in ASPEN Plus should be on a dry basis. The additional input needed is the heat of combustion that is specified as "HCOMB" in Properties > Methods > Parameters > Pure Components > Heat. The inbuilt example of solid2.bkp from ASPEN Plus Examples "Getting Started with Solids" was used as a starting point to set up the simulation. The gasification reaction is modeled by using two hypothetical reactors in series. This is done to convert the non-conventional components to elemental composition in the first reactor (DECOMP in ASPEN Plus example). The conversion of the elements to the final syngas composition is carried out in the second reactor and is controlled with the help of an RYIELD block (Yield reactor model in ASPEN Plus). Since both the reactors are simulating a single reactor, they are heat-connected with a heat stream going from the first to the second reactor, which removes a degree of freedom but closely resembles the actual conditions of the gasification reactor. The second reactor is modeled as an RYIELD block where the gasification reaction information is the input.

The mass yields are calculated based on the experimental set of data and are shown in **Table S1**. To make the model more dynamic to syngas yields, a simple formula was deduced so that the gas yields change as per the syngas yields. This was important since the syngas yield is a parameter that was varied later in the sensitivity analyses, and it is hence essential that the gas/solid distributions change as the syngas yield is changed. The yield of each gas is an input in the ASPEN model. The yield is defined as mass of the gas produced per kg of total input (MPW + steam). Based on this definition, and the mass balance shown in **Table S1**, the following equation is obtained. The equation is used in the ASPEN model to calculate yield of a specific gas in the model based on the experimental data:

$$\mathbf{Y_{gas}} = \frac{0.00044642 \times \mathrm{MW_{gas}} \times \dot{\mathrm{m}}_{\mathrm{MPW}} \times \mathbf{Y_{syngas}} \times \mathrm{X_{gas}}}{(\dot{\mathrm{m}}_{\mathrm{MPW}} + \dot{\mathrm{m}}_{\mathrm{Steam}})}$$

$\mathrm{MW_{gas}} \rightarrow \frac{\mathrm{g}}{\mathrm{mol}}$

$\dot{\mathrm{m}}_{\mathrm{MPW}} + \dot{\mathrm{m}}_{\mathrm{Steam}} \rightarrow \mathrm{kg/h}$

$\mathrm{Y_{syngas}} \rightarrow$ Syngas yield in $\mathrm{Nm^3/kg}$

$\mathrm{X_{gas}} \rightarrow$ Composition of gas in gasifier outlet in %

Syngas yield for gasification process on mass basis. To compare the performance of different gasification feedstocks using different plastics, it is important to use a consistent scale to measure the efficiency of gasification processes. Reports in the literature[6,14] sometimes use Nm^3/kg (syngas volumetric flow/solid feedstock) for comparison of different processes. The problem with using these units is that some gasification processes may produce more light hydrocarbons (CH_4 and C_2H_6) than CO and H_2, which reduces the volumetric flow rate of syngas (by a reduction in the number of moles). CH_4 or C_2H_6 in the gasifier outlet does not necessarily affect the process economics. Because there are two more opportunities downstream of the gasifier for reforming (tar reforming and steam reforming), the hydrocarbons are eventually converted to syngas. Hence, the mass yield approach (kg syngas/kg solid feedstock) that is used in this study normalizes the effect of product distribution of different gases in the syngas and is a better metric than Nm^3/kg to compare different gasification technologies. Any reduction in the mass yield indicates the loss of solid feedstock as a solid residue which directly impacts process economics. The H_2/CO ratio in the syngas should also be specified when comparing different gasification processes.

Table S22. *Energy balance for gasification reactor*

Stream enthalpy in, Gcal/h	Stream enthalpy out, Gcal/h
Feed stream, -10.88	Products, -44.4
Steam, -62.66	
Total In: -73.54	Total out: -44.4

$$\Delta \mathrm{H_{r,gasification}} = \mathrm{H_{products}} - \mathrm{H_{reactants}} = -44.38 + 73.54 = 29.14\ \mathrm{Gcal/h}$$

Energy balance around gasifier. Table S22 shows the energy balance around the gasification reactor. The enthalpy, ΔH of all the reactions happening inside the reactor is +29.14 Gcal/h (12.19 MJ/kg plastic feed). The positive value is expected since mixed plastic waste gasification is endothermic and needs energy to proceed, and this is the same energy that is input into the gasifier by the combustion reactor. The heat of reaction value can be compared to the heat of reactions for other steam reforming of hydrocarbon reactions (methane 12.89 MJ/kg, ethane 11.55 MJ/kg, propane 9.86 MJ/kg).

Syngas preparation. After particulate removal, syngas conditioning involves sulfur removal and steam methane reforming, as shown in **Fig. S14**. The sulfur content in MPW is 50 ppm. Hence, bulk sulfur removal was not necessary. Only one ZnO bed has been considered in the design, which can reduce the sulfur in the syngas down to 1 ppm[16].

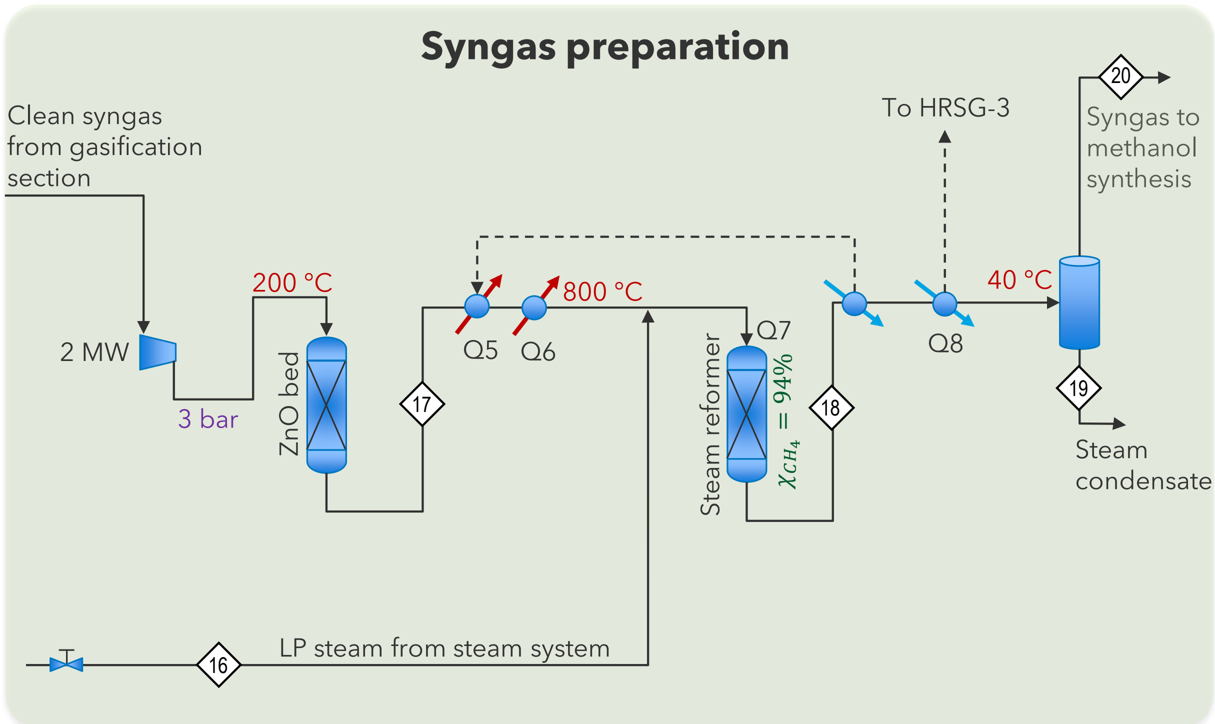

Fig. S14. *Syngas preparation PFD for MPW-Methanol process.* Refer to Table S12 for heat transfer duties.

After sulfur removal, the syngas is passed over a Ni catalyst in a fixed bed steam reformer. The operating pressure of the steam reformer is 3 bar, lower than the conventional 20-25 bar used in industry[4] to enhance methane conversion rates in the reformer. In our conceptual process design, the steam reformer comes after the gasification reactor and tar reformer. Hence the syngas already has a high concentration of CO and H_2 (almost 80%). Natural gas steam reforming systems do not face this problem as the feed gas to the reformer contains no CO and H_2 and the equilibrium easily shifts forward resulting in very high methane conversions. But in the MPW-Methanol process, the reformer feed already contains high concentrations of CO and H_2 and at 20 bar pressure, the equilibrium shifts backward in the steam reforming reaction due to Le Chatelier's principle, thereby lowering methane conversion rates. Hence, a lower pressure is more suitable in this case which is also in line with patent literature[6] on the MSW gasification process considered in this study. However, the choice of a lower pressure comes at the cost of an increase in the compression duty of the make-up gas compressor upstream of the methanol synthesis reactor.

Methanol synthesis loop. After reforming, the syngas is injected into the 'Synthesis loop'. The make-up gas compressor increases the pressure from about 3 bar to 80 bar as shown in **Fig. S15**. The methanol-to-dimethyl ether (DME) reaction has been added to simulate the production of the lighter side products. The methanol process uses a recycle loop configuration[12] due to low single-pass conversions of about 40%. A recycle gas compressor (RGC) maintains circulation flow of the unreacted syngas within the loop. A purge stream ensures there is no accumulation of inerts in the loop. The main inerts in the loop are unconverted methane and ethane. The purge fraction was set at 15%.

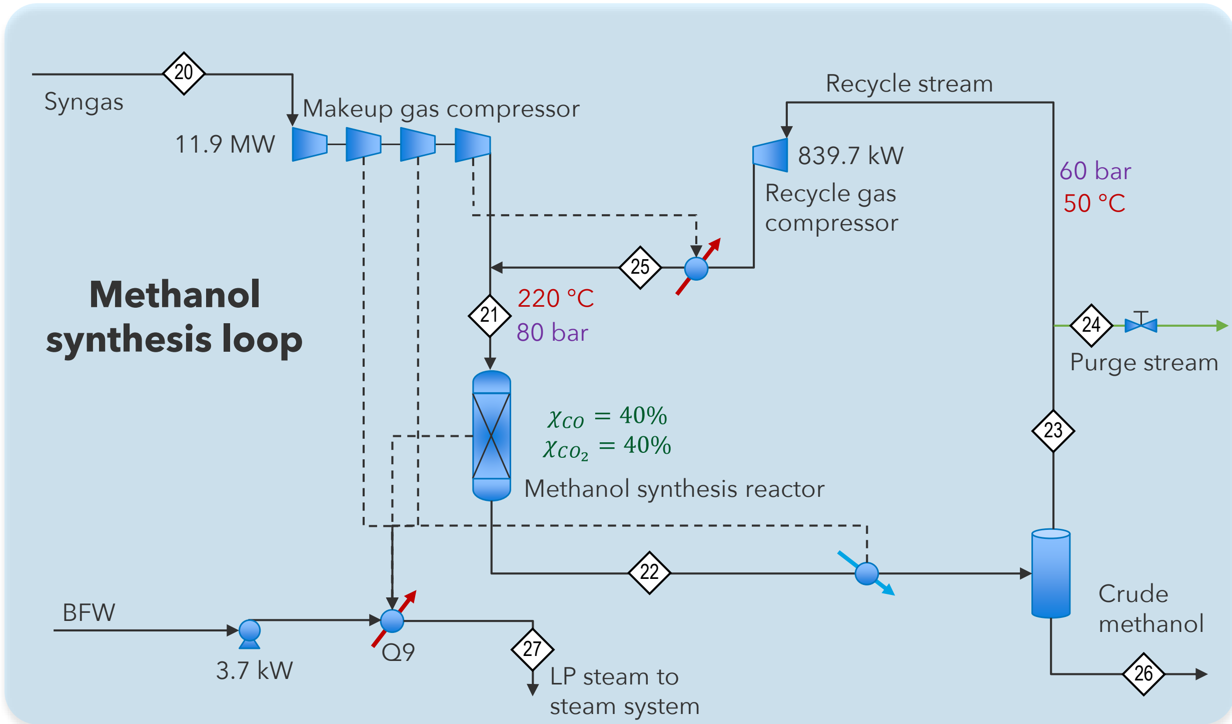

Fig. S15. *Methanol synthesis loop PFD for MPW-Methanol process*

A sensitivity analysis was performed to study the effect of the purge split fraction on the mole fraction of the inlet stream. As shown in **Fig. S16**, as the purge fraction is increased, the amount of purge stream increases resulting in a slight increase in reactant concentration in the MeOH reactor feed (green line). This is good from a kinetic perspective, but the negative impact is in the decrease of final methanol product production. Due to the material loss of reactants from the loop (CO, CO_2, H_2), methanol production decreases. However, for all cases, the reactants' mole fraction is consistently above 90%. Hence, a different approach was used to fix the split ratio.

The purge stream is used as fuel in the combustion reactor in gasification section. Hence, a low split fraction indicates the need to use external fuel to be used in the combustion reactor. On the other hand, a high split fraction results in reduction of methanol production rate (indicated by the blue line). Hence, an intermediate value of 0.15 was chosen for the split fraction. At this value of split fraction, the methanol production rate is about 354 MT/D. The split fraction will be a sensitivity variable that will be varied to study the impact on the overall process and MSP of methanol product.

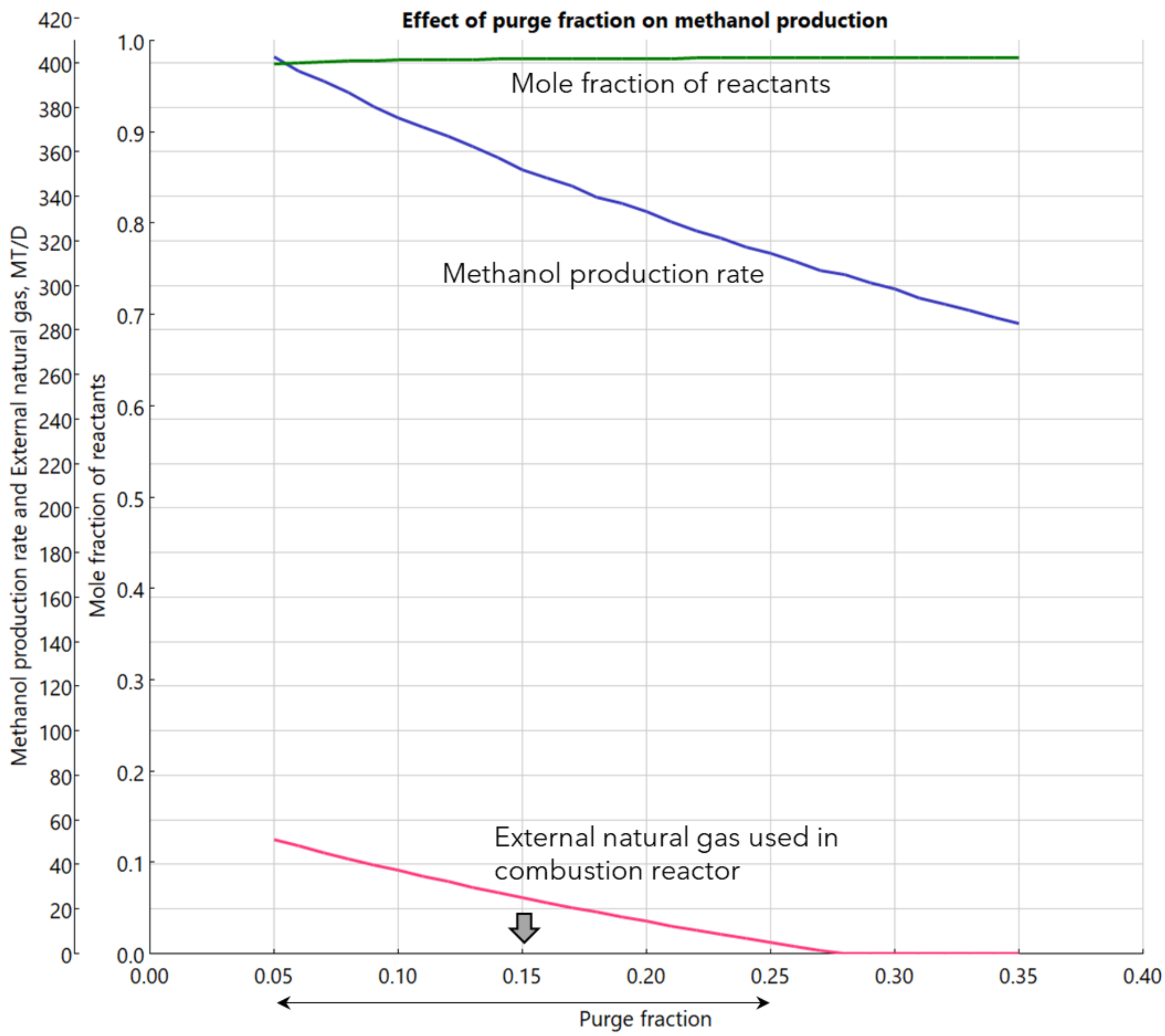

Fig. S16. *Effect of purge split fraction on import natural gas*

Methanol distillation. The distillation section is designed based on the purity required for different applications[17]: fuel, solvent, or chemical synthesis. In our design, we have considered chemical synthesis application and set the desired purity to 99.8%. Accordingly, two distillation columns were needed for the separation as shown in **Fig. S17**. For very high methanol capacities above 5000 MT/D, three or four columns in series may be required[18].

The boiling point of methanol is 65 °C, an intermediate value between the side products like DME (b.p. -24 °C) and water (100 °C). The separation section helps increase the purity from about 75% to 99.8% by removing DME as the top product in the first column and water as the product in the second column. Two design specs have been used in the ASPEN Plus model to ensure the required product quality for methanol and maximize methanol recovery. In the first distillation column, the pressure is lowered to 11 bar and all gases from the top, which mainly contain methanol, H_2, and DME are routed to the gasification section for combustion and energy recovery. The bottom product in the first column is distilled in a second column and 99.8% methanol is recovered from the top of the column. The water collected at the bottom of the heavy ends distillation column has some methanol in it and is routed to wastewater treatment.

After the base case simulation was completed, the simulation results were imported into a custom-built excel sheet where the equipment was sized, and their operating expenditures (OPEX) and capital expenditures (CAPEX) were estimated. Once the techno-economic analysis (TEA) was complete, a minimum selling price (MSP) for the product was estimated based on discounted cash flow rate of return (DCFROR) analysis. Economic assumptions include a 2016 cost year basis, 90% onstream time, 10% IRR, and 21% tax rate.

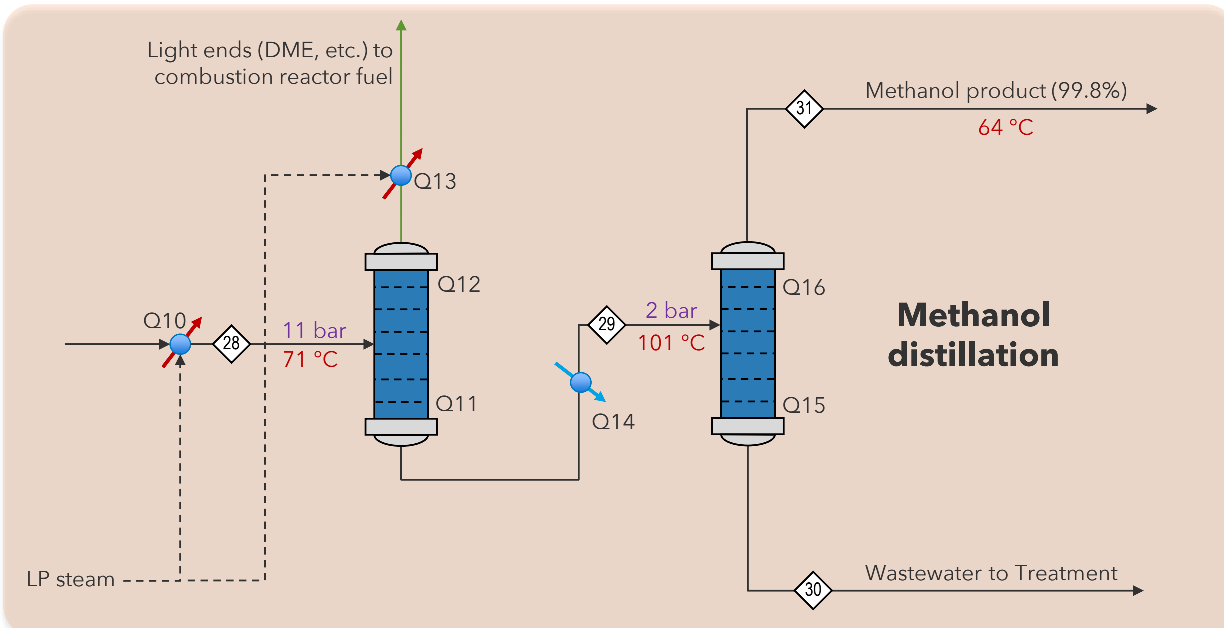

Fig. S17. *Methanol distillation PFD for MPW-Methanol process*

Fig. S10 tracks the mass of carbon throughout the process and where the carbon ends up in the product. The main loss of carbon in the gasification section is the loss of carbon as char which is burnt to produce energy. The remaining carbon is converted to syngas and is processed in the methanol synthesis section. The purge results in a large loss of carbon from the recycle loop. About 63% of the initial carbon ends up in methanol. Carbon balance tells only part of the story as the carbon atom picks up the oxygen molecule in methanol formation resulting in enhancing the mass yield. The loss of carbon is compensated by the oxygen molecule and methanol mass yield is ~1.47 kg methanol/kg mixed plastic waste feedstock.

Combustion v/s chemical synthesis pathways for MPW conversion. Waste plastics are chemically just hydrocarbons and combusting them should provide energy. Power generation was not considered since chemical synthesis produces a higher value of products than electricity production.[19] A waste to energy plant combusting waste plastics will definitely see an increase in the electricity generation since calorific value of plastics is high. However, from an economic perspective, combusting waste plastic to generate electricity is not attractive. A simple calculation can ascertain that the economic value of chemical products from 1 kg of MPW is much more than that obtained from electricity generation by combustion. Based on plastics chemical composition in **Table 2**, combustion of 1 kg of MPW releases 43.4 MJ/kg (12.06 kWh/kg). Since WTE plants have efficiencies of 22-25%,[20] assuming 23.5% efficiency and electricity sale price of 4.37 ¢/kWh, revenue by selling electricity is 11.3 ¢/kg. For comparison, the conversion of waste plastics to methanol can be summarized as ($–CH_2–$ $+H_2O \rightarrow CH_3OH$)

Based on stoichiometry, 1 kg of MPW can produce 2.29 kg methanol (from 32/14, i.e., the molecular weight of methanol/molecular weight of reactant unit, CH_2). The methanol market price is $0.30/kg; hence revenue is $0.71/kg MPW. This assumes 100% yield, but actual mass yield as reported in **Table 3** is 1.47 kg methanol/kg MPW. Hence, actual revenue reduces to 44.1 ¢/kg MPW. Therefore, the economic value of products produced by 1 kg of MPW could be about 4 times more in chemicals produced by synthesis pathways, like methanol than from energy generation by combustion.

Table S23. *Stream summary for MPW-Methanol process.* Detailed PFDs for MPW-Methanol process are shown in **Fig. S13 - S15, S17**.

Stream no. →		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
Temperature, C		25.0	190.6	30.0	25.0	108.6	46.6	835.1	880.0	835.1	835.1	835.1	890.0	40	40.0	40.0
Pressure, bar		1	3	1	1	1	1	1	1	1	1	1	1	1	1	1
Mass vapor fraction		0	1	1	1	1	1	0	1	0.97	1	0	1	0	0	1
Mass liquid fraction		0	0	0	0	0	0	0	0	0	0	0	0	1	1	0
Mass solid fraction		1	0	0	0	0	0	1	0	0.03	0	1	0	0	0	0
MT/D	Total mass flow	240.0	480.0	1858.1	25.2	34.6	120.1	22.5	2060.6	720.0	695.5	2.0	695.5	222.5	0.8	472.1
	H_2O	-	480.0	-	-	1.3	0.2	-	239.4	346.7	346.7	-	264.4	222.5	-	41.8
	H_2	-	-	-	-	0.0	16.7	-	-	21.2	21.2	-	40.4	-	-	40.4
	CO	-	-	-	-	0.2	39.9	-	-	137.9	137.9	-	265.9	-	-	265.8
	CO_2	-	-	-	-	11.0	58.4	-	323.8	49.3	49.3	-	49.3	-	-	49.2
	CH_4	-	-	-	25.2	-	1.4	-	-	61.0	61.0	-	48.8	-	-	48.8
	C_2H_4	-	-	-	-	-	-	-	-	50.2	50.2	-	25.1	-	-	25.1
	C_2H_6	-	-	-	-	-	-	-	-	3.8	3.8	-	0.4	-	-	0.4
	C_3H_8	-	-	-	-	-	-	-	-	9.9	9.9	-	0.5	-	-	0.5
	$C_{10}H_8$	-	-	-	-	-	-	-	-	15.5	15.5	-	0.8	-	0.8	-
	N_2	-	-	1425.3	-	-	-	-	1425.3	-	-	-	-	-	-	-
	O_2	-	-	432.8	-	-	-	-	72.1	-	-	-	-	-	-	-
	C	-	-	-	-	-	-	22.5	-	22.5	-	-	-	-	-	-
	Ash	-	-	-	-	-	-	-	-	0.2	-	0.2	-	-	-	-
	Char2	-	-	-	-	-	-	-	-	1.7	-	1.7	-	-	-	-
	MPW	240.0	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	CH_3OH	-	-	-	-	21.4	2.9	-	-	-	-	-	-	-	-	-
	CH_3-O-CH_3	-	-	-	-	0.5	0.6	-	-	-	-	-	-	-	-	-

Table S24. *Stream summary for MPW-Methanol process (contd.).* Detailed PFDs for MPW-Methanol process are shown in **Fig. S13 - S15, S17**.

	Stream no. →	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31
	Temperature, °C	190.6	187.5	800.0	40.0	40.0	226.4	220.0	50.0	50.0	232.0	50.0	200.0	71.0	101.0	88.8	63.8
	Pressure, bar	3	3	3	3	3	80	80	60	60	80	60	3	11	2	1	1
	Mass vapor fraction	1	1	1	0	1	1	1	1	1	1	0	1	0	1	0	0
	Mass liquid fraction	0	0	0	1	0	0	0	0	0	0	1	0	1	0	1	1
	Mass solid fraction	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
MT/D	Total mass flow	795.3	472.1	1267.4	611.8	655.6	1336.4	1336.4	800.9	120.1	680.8	535.4	960.0	535.4	500.8	147.1	353.8
	H_2O	795.3	41.8	633.1	611.8	21.2	22.6	132.3	1.6	0.2	1.4	130.7	960.0	130.7	129.3	128.7	0.7
	H_2	-	40.4	78.9	-	78.9	173.4	111.2	111.1	16.7	94.4	-	-	-	-	-	-
	CO	-	265.8	217.8	-	217.8	444.1	266.5	266.3	39.9	226.3	0.2	-	0.2	-	-	-
	CO_2	-	49.2	336.2	-	336.2	667.0	400.2	389.2	58.4	330.8	11.0	-	11.0	-	-	-
	CH_4	-	48.8	1.4	-	1.4	9.1	9.1	9.1	1.4	7.7	-	-	-	-	-	-
	C_2H_4	-	25.1	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	C_2H_6	-	0.4	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	C_3H_8	-	0.5	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	$C_{10}H_8$	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	N_2	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	O_2	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	C	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	Ash	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	Char2	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	MPW	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	CH_3OH	-	-	-	-	-	16.5	412.3	19.4	2.9	16.5	392.9	-	392.9	371.5	18.4	353.1
	CH_3-O-CH_3	-	-	-	-	-	3.7	4.8	4.3	0.6	3.7	0.5	-	0.5	-	-	-

S4. MSW-Methanol process

Most process details for the MSW-Methanol process remain the same as explained in the MPW-Methanol process, and any deviations from it are described below.

A direct gasification design is used for the MSW-Methanol process as shown in **Fig. S18**. The energy to drive the gasification reactions is supplied directly within the gasifier and hence oxygen is required as input in the MSW gasifier. The gasifier design and output syngas composition are based on a commercial patent – Production and conditioning of synthesis gas obtained from biomass.[6] The solid residue from the gasifier bottom in MSW gasification is not solely char, as was in the case of MPW, but could contain non-volatilized components. Hence it is disposed and not combusted eliminating the use of a char combustion reactor.

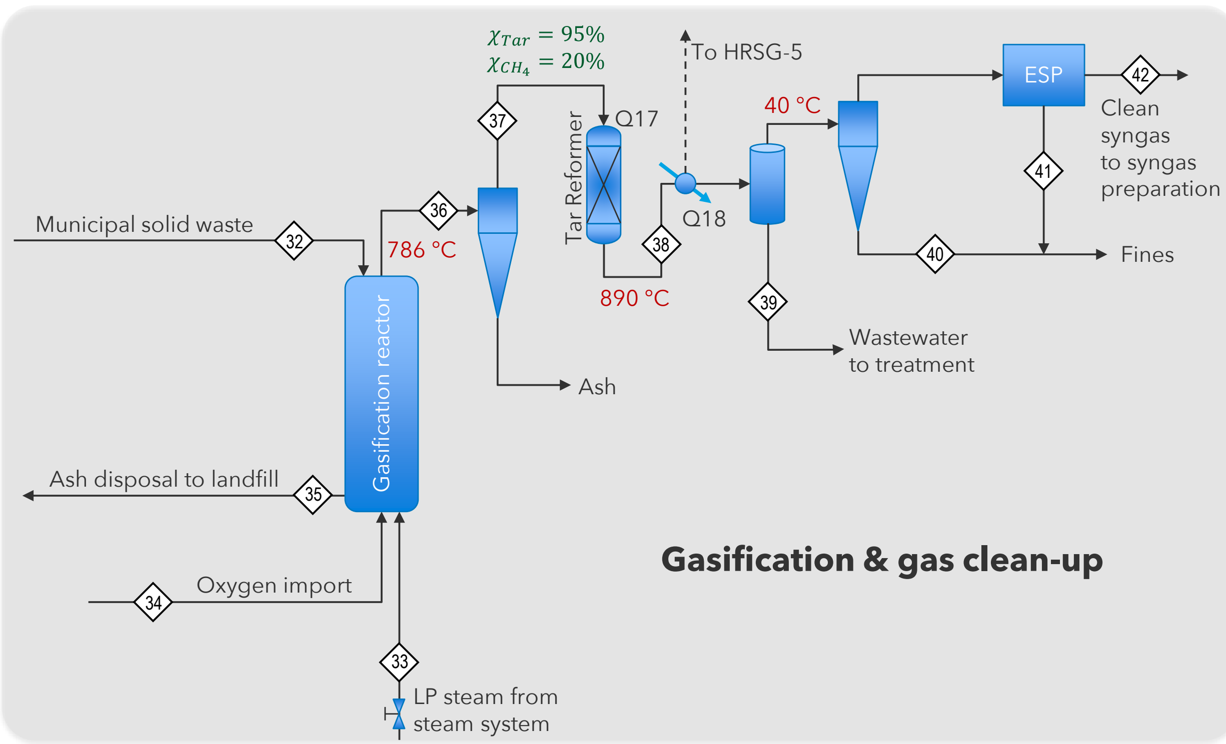

Fig. S18. *Gasification and gas clean-up for MSW-Methanol process*

The syngas yield for MSW-related processes is lower than that for MPW processes. The syngas yield is 0.8-0.9 Nm^3/kg for a process involving MSW gasification[21] compared to 2.1 Nm^3/kg that was used for MPW gasification[1]. Pure oxygen gasification of MSW has a lower dry gas yield of 0.7 Nm^3/kg based on patent literature[22]. The important parameter, H_2/CO ratio is 0.58, much lower than 2.1 for MPW gasification. The H_2/CO ratio is boosted by reactions in the tar reformer and steam reformer to 2, which is needed for methanol synthesis. The CO_2 composition in syngas after MSW-Gasification is 41%, much higher than the 5% in the MPW case. This can be attributed to the use of oxygen in MSW gasification that produces CO_2 and more importantly to the MSW composition which has 29% oxygen. This results in loss of carbon as CO_2 and impacts carbon efficiency negatively.

S/C ratio in steam reformer for MSW-Methanol process. The S/C ratio in the steam reformer for all processes was assumed to be 3, except the MSW-Methanol process, where the S/C for the base case was set at 1.3 as per patent literature for MSW gasification.[6] The reasoning follows from the reactions occurring at the downstream methanol synthesis reactor. Specifically, for the MSW-Methanol process, a high S/C ratio in the steam reformer results in a high CO_2 concentration in the reformer outlet due to the WGS reaction effect. The CO_2 removal unit before the methanol synthesis reactor removes CO_2 from the syngas to maintain a low CO_2 mole fraction at the reactor inlet to enhance methanol formation rates. Hence, a high S/C ratio at the steam reformer results in loss of the reactant CO as CO_2. The loss of CO affects methanol yield and ultimately hurts the methanol MSP, which is evident in by the large impact of the S/C ratio in the sensitivity analysis tornado plot as shown in **Fig. S4**. Such an effect is not seen in the other processes in this study; the MPW-Methanol process does not have a CO_2 removal unit. In the MPW-Hydrogen and MSW-Hydrogen processes, the loss of CO as CO_2 does not adversely affect process economics as the final product does not contain any carbon. Therefore, the S/C ratio for the MSW-Methanol process was fixed at 1.3.

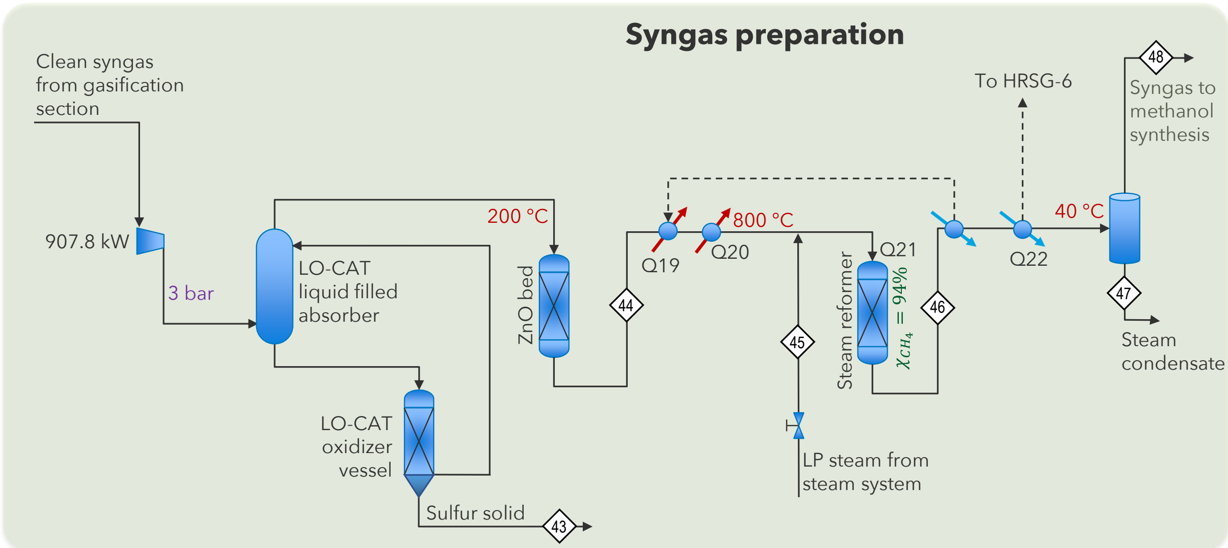

Fig. S19. *Syngas preparation PFD for MSW-Methanol process*

The sulfur content in MSW is 0.3% by wt. corresponding to 3000 ppmw. Hence, bulk sulfur removal is necessary which is accomplished by a liquid oxidation catalyst in the LO-CAT system. It is a liquid redox system that uses a chelated iron solution to reduce H_2S to elemental sulfur[23]. The sulfur content reduces to about 50 ppm and the remainder is removed by a ZnO bed. The syngas preparation PFD for MSW-Methanol process is shown in **Fig. S19**.

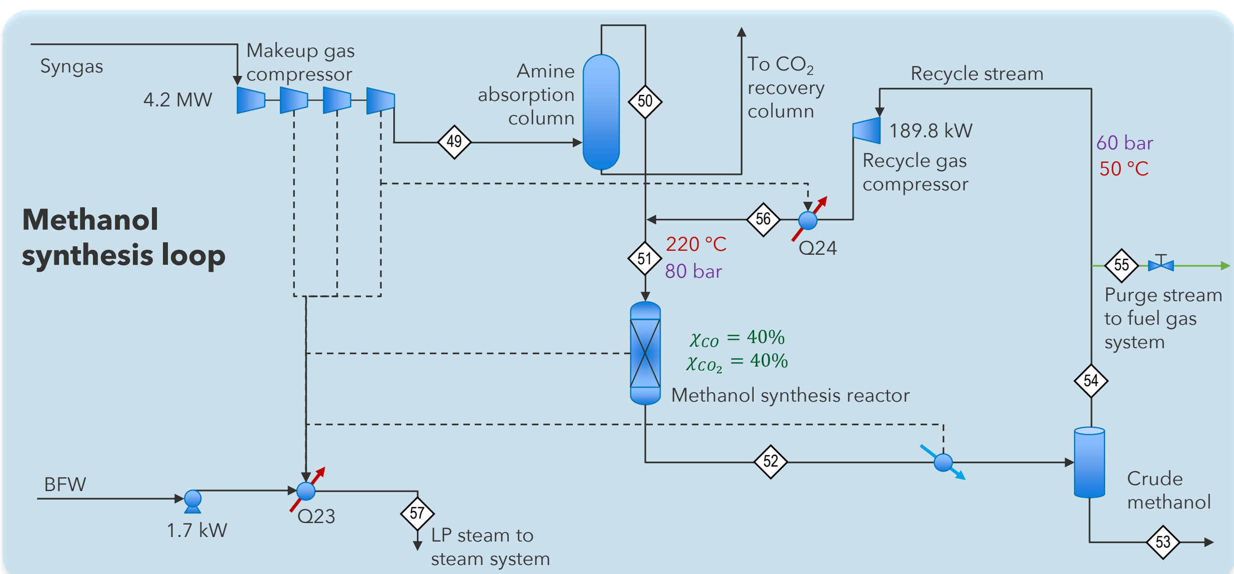

Fig. S20. *Methanol synthesis PFD for MSW-Methanol process*

The CO_2 content in the syngas after MSW gasification and reforming steps is high at 21%. Though CO_2 is a reactant in the methanol synthesis reaction, it is usually limited to about 7.5% at the reactor inlet to maximize methanol production rates based on kinetic studies[24]. To accomplish this, a CO_2 recovery unit was added to remove as much CO_2 as necessary to ensure a mole fraction of 0.075 for CO_2 at the methanol synthesis reactor inlet. The PFD for methanol synthesis and distillation are shown in **Fig. S20** and **Fig. S21** respectively. The MSW gasifier does not have a combustion reactor, and hence the purge gas and light ends from the first distillation column is routed as fuel to reformer furnaces as shown in **Fig. S22**.

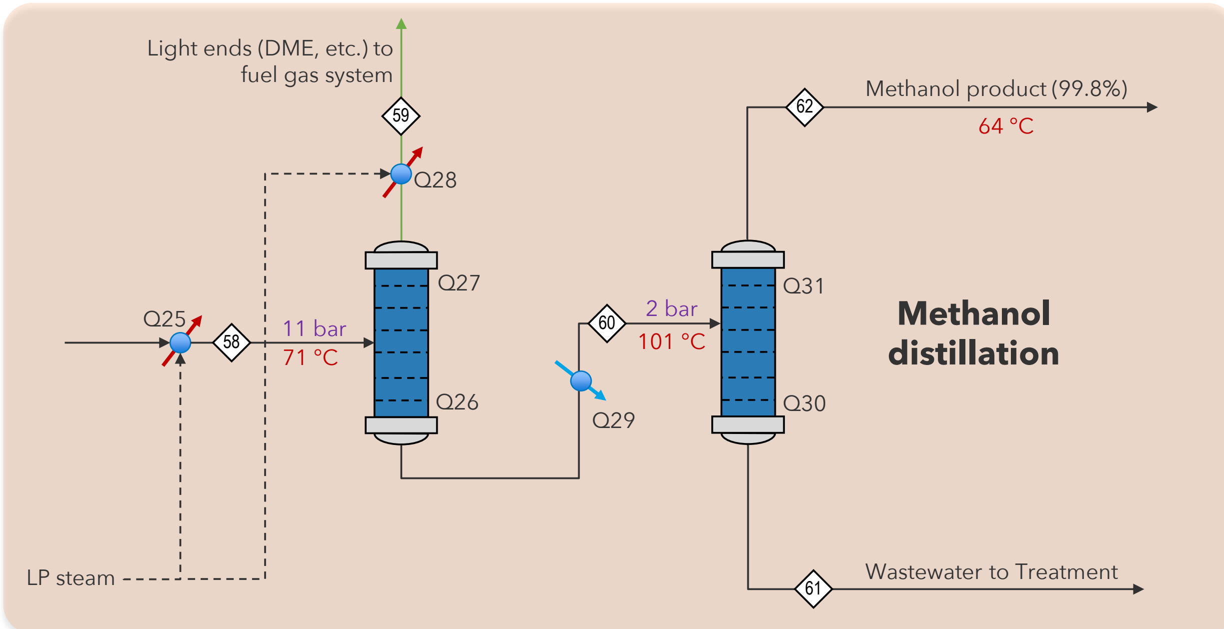

Fig. S21. *Methanol distillation PFD for MSW-Methanol process*

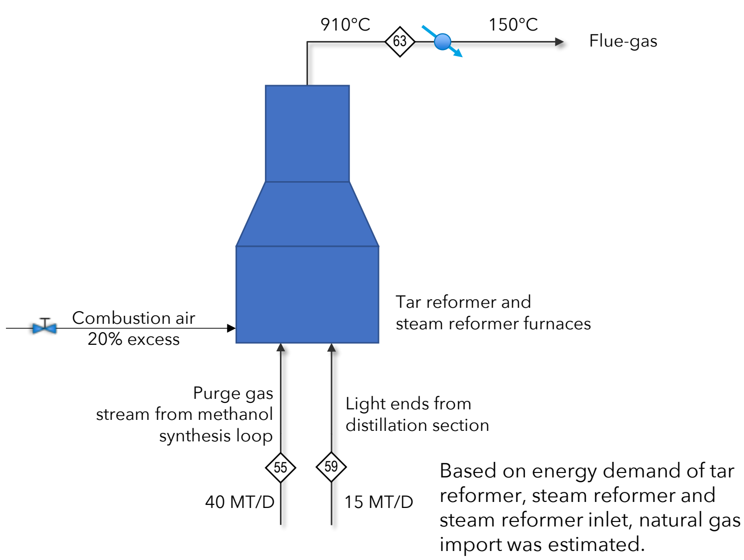

Fig. S22. *Fuel-gas system for MSW-Methanol process*

Table S25. *Stream summary of MSW-Methanol process.* Detailed PFDs for MSW-Methanol process are shown in **Fig. S18 – S22**.

	Stream no. →	32	33	34	35	36	37	38	39	40	41	42	43	44	45	46	47	48
	Temperature, °C	25.0	152.5	25.0	785.7	785.7	785.7	890.0	40.0	40.0	40.0	40.0	45.0	183.2	152.5	800.0	40.0	40.0
	Pressure, bar	1	3	1	1	1	1	1	1	1	1	1	1	3	3	3	3	3
	Mass vapor fraction	0	1	1	0	0.86	1	1	0	1	0	1	0.68	1	1	1	0	1
	Mass liquid fraction	0	0	0	0	0.00	0	0	1	0	1	0	0.32	0	0	0	1	0
	Mass solid fraction	1	0	0	1	0.14	0	0	0	0	0	0	0	0	0	0	0	0
MT/D	**Total mass flow**	240.00	105.97	50.03	56.80	396.00	339.20	339.20	32.49	0.03	0.33	306.36	0.15	306.23	174.05	480.28	155.52	324.76
	H_2O	-	105.97	-	-	106.03	106.03	51.79	32.48	-	-	19.31	0.01	19.31	174.05	162.97	155.51	7.46
	H_2	-	-	-	-	1.74	1.74	15.62	-	-	-	15.62	-	15.62	-	21.79	-	21.79
	CO	-	-	-	-	41.53	41.53	125.86	-	0.01	-	125.85	-	125.85	-	120.17	-	120.17
	CO_2	-	-	-	-	131.49	131.49	131.49	-	0.01	-	131.48	0.03	131.48	-	173.05	-	173.05
	CH_4	-	-	-	-	11.60	11.60	9.28	-	-	-	9.28	-	9.28	-	0.64	-	0.64
	C_2H_4	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	C_2H_6	-	-	-	-	22.40	22.40	2.24	-	-	-	2.24	-	2.24	-	-	-	-
	C3H6	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	C_3H_8	-	-	-	-	16.01	16.01	0.80	-	-	-	0.80	-	0.80	-	-	-	-
	$C_{10}H_8$	-	-	-	-	6.62	6.62	0.33	-	-	0.33	-	-	-	-	-	-	-
	N_2	-	-	-	-	1.65	1.65	1.65	-	-	-	1.65	0.07	1.65	-	1.65	-	1.65
	O_2	-	-	50.03	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	S	-	-	-	-	-	-	-	-	-	-	-	0.04	-	-	-	-	-
	Ash	-	-	-	43.92	43.92	-	-	-	-	-	-	-	-	-	-	-	-
	Char2	-	-	-	12.88	12.88	-	-	-	-	-	-	-	-	-	-	-	-
	MSW	240.00	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	COS	-	-	-	-	0.04	0.04	0.04	-	-	-	0.04	-	-	-	-	-	-
	NH3	-	-	-	-	0.07	0.07	0.07	-	-	-	0.07	-	-	-	0.01	-	-
	H2S	-	-	-	-	0.02	0.02	0.02	-	-	-	0.02	-	-	-	-	-	-

Table S26. *Stream summary of MSW-Methanol process (contd.).* Detailed PFDs for MSW-Methanol process are shown in **Fig. S18 – S22**.

	Stream no. →	49	50	51	52	53	54	55	56	57	58	59	60	61	62	63
	Temperature, °C	220.0	220.0	219.7	220.0	50.0	50.0	50.0	219.6	152.7	71.0	103.3	101.0	85.5	63.8	910.0
	Pressure, bar	80	80	80	80	60	60	60	80	3	11	9	2	1	1	1
	Mass vapor fraction	1	1	1	1	0	1	1	1	1	0	1	1	0	0	1
	Mass liquid fraction	0	0	0	0	1	0	0	0	0	1	0	0	1	1	0
	Mass solid fraction	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
MT/D	Total Mass flow	324.76	203.79	426.55	426.55	164.48	262.08	39.31	222.76	337.99	164.48	16.11	148.36	33.90	114.46	382.54
	H_2O	7.46	7.46	7.72	28.25	27.95	0.30	0.05	0.26	337.99	27.95	0.18	27.77	27.55	0.22	40.26
	H_2	21.79	21.79	34.04	14.42	0.01	14.41	2.16	12.25	-	0.01	0.01	-	-	-	-
	CO	120.17	120.17	244.81	146.88	0.25	146.63	21.99	124.64	-	0.25	0.25	-	-	-	-
	CO_2	173.05	52.08	100.45	60.27	3.36	56.91	8.54	48.38	-	3.36	3.36	-	-	-	77.01
	CH_4	0.64	0.64	4.08	4.08	0.03	4.04	0.61	3.44	-	0.03	0.03	-	-	-	-
	C_2H_4	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	C_2H_6	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	C_3H_6	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	C_3H_8	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	N_2	1.65	1.65	10.76	10.76	0.04	10.73	1.61	9.12	-	0.04	0.04	-	-	-	252.57
	O_2	-	-	-	-	-	-	-	-	-	-	-	-	-	-	12.69
	CH_3OH	-	-	3.98	130.73	126.04	4.69	0.70	3.98	-	126.04	5.45	120.59	6.35	114.24	-
	CH_3-O-CH_3	-	-	20.70	31.14	6.79	24.36	3.65	20.70	-	6.79	6.79	-	-	-	-
	NH_3	-	-	0.01	0.01	-	-	-	-	-	-	-	-	-	-	-

Capital cost and annual operating cost estimation. For the MSW-Methanol process with MSW feed capacity of 240 MT/D, the total capital investment (TCI) is \$79M and the annual operating cost is \$11M. Based on a previous report,[19] the total capital cost estimation for the same process and same MSW processing capacity was \$70M (scaled based on the six-tenth rule). The cost breakdown of CAPEX and OPEX is shown in Fig. . For capital costs, the breakdown is similar to the MPW-Methanol process; the combined contribution of gasification and syngas preparation sections to the overall CAPEX is slightly higher in MSW-Methanol process when compared to MPW-Methanol process (34% vs 29%). The increase can be attributed to the additional sulfur removal equipment (LO-CAT) used in the MSW process. In operational costs, the primary material input is oxygen. The OPEX is much lower in the MSW-Methanol process compared to the MPW-Methanol process since the MSW feedstock cost is assumed to be zero in the base case.

Sensitivity analysis on key cost drivers for hydrogen production. A similar sensitivity analysis was carried out for the MSW-Methanol process and the results are shown in **Fig. S4**. It is interesting to note that syngas yield, which was a very critical parameter in MPW-Methanol is not so for the MSW-Methanol process. Reducing the syngas yield by 50% in the MPW process resulted in an 86% increase in methanol MSP. However, reducing the syngas yield by 50% in the MSW-methanol case results in a 28% increase in methanol MSP (extrapolated linearly). The larger impact of syngas yield on the methanol MSP is due to the feedstock cost and waste disposal costs. The mixed plastic waste is available for a high cost (\$0.60/kg). Any reduction in syngas yield has two negative effects on process economics – (1) reduction in product yield and (2) increase in solid residue from the gasifier. Solid waste disposal is assumed at the US average rate of \$59/MT.[7] The same effect is not seen for MSW as the feedstock is considered zero cost for the base case. Hence, though a reduction in syngas yield increases MSP, other parameters like plant capacity, IRR, and tipping fee have a larger impact. Hence, for the same downstream product, a change in feedstock changes the relative impact of different variables on the product MSP.

Of all the variables studied, the plant capacity has the largest impact on the methanol MSP from the MSW-Methanol process. The MSW-Methanol process is sensitive to economies of scale and a larger plant will exhibit better process economics. The process-relevant variables of methanol loop purge fraction and single-pass conversion in the methanol reactor impact the methanol MSP from -9% to +12% for the ranges studied. Variations in the methanol side-products % (methanol to DME conversion) have a smaller impact on the MSP with their effect being only ±4% from the base case. Since oxygen is fed to the gasifier, the effect of including the air separation unit (ASU) CAPEX was assessed. The MSP increases by 8% if the ASU CAPEX is included in the capital costs of the MSW-Methanol process. The capital costs were taken from literature[25] and scaled for the current O_2 feed requirement.

The tipping fee is the disposal fee charged by the owner or operator of a landfill site, and it varies widely throughout different regions in the US. Hence, when using MSW as a feedstock, the plant operator obtains the feedstock as well as the tipping fee from municipalities disposing the MSW. The US national average tipping fee is \$59/MT.[7] A \$20/MT cost was added to account for the conversion of MSW to refuse-derived fuel (RDF), which is the MSW in pelletized form input to the gasifier.[8] Considering the additional income from the tipping fee for the MSW-Methanol process, the MSP of methanol reduces to \$0.49/kg. The average tipping fee in the Pacific region is \$79/MT,[7] and the corresponding methanol MSP reduces further to \$0.44/kg. Hence, securing long-term tipping fee agreements with city municipalities is one of the key requirements for the economic viability of MSW feedstock-based plants.

Varying the IRR from 5% to 15% impacts the methanol MSP by as much as ±16% indicating a major effect. Similarly, varying TCI also affects the methanol MSP by -9% to +18% in the range studied. The impact of the financial variables is more prominent in the MSW-Methanol process compared to the MPW-Methanol process. In the MPW-Methanol process, the waste plastic feedstock results in a large annual operating cost of \$62M compared to its capital cost of \$75M. On the other hand, the MSW-Methanol has lower annual operating costs of \$11M when compared to its capital cost of \$40M. Hence, changes in capital costs affect the process economics more than the MPW-Methanol process.

S5. MPW-Hydrogen process

The gasification and syngas preparation sections for the MPW-Hydrogen process remain identical as in the MPW-Methanol process, which was described in detail earlier. The modification is downstream of syngas preparation which is described here.

Water-gas-shift (WGS). The syngas after syngas preparation has a CO concentration of about 14%. Since hydrogen is the final desired product, a water-gas-shift reactor is used to convert the CO to H_2 by the WGS reaction. The CO conversion was fixed at 90%[26] using an RSTOIC block in ASPEN Plus. The PFD for WGS of the MPW-Hydrogen process is shown in **Fig. S23**.

Pressure swing adsorption (PSA). The PSA adsorbents are selected based on the composition of the impurities and are usually a combination of silica gel, alumina, activated carbon, and zeolite[27]. Most of the hydrogen passes without being adsorbed though there are some losses by adsorption. A cyclic process is used with each bed in either adsorption, depressurization, or purge mode. An elaborate control system is used to automate the control valves in a PSA unit. The PSA PFD for MPW-Hydrogen process is shown in **Fig. S24**.

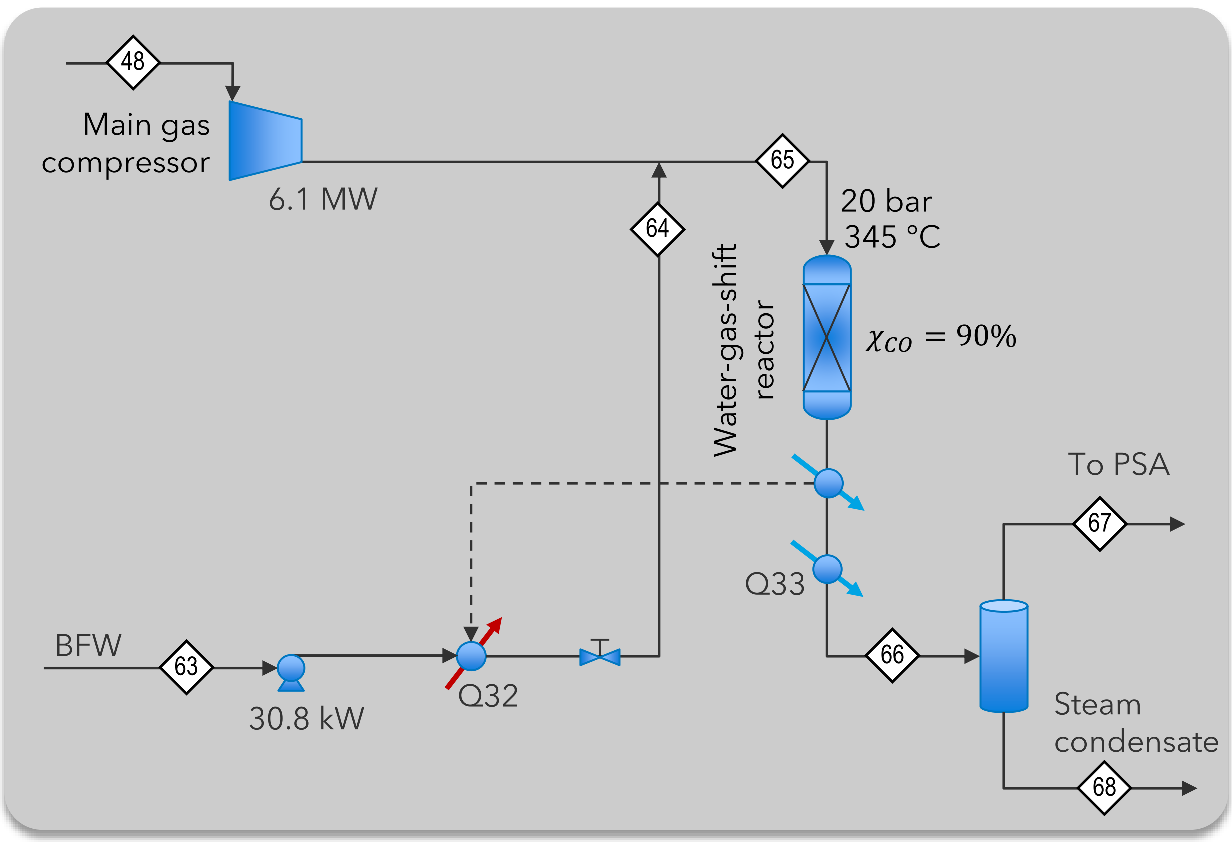

Fig. S23. *WGS PFD for MPW-Hydrogen process.*

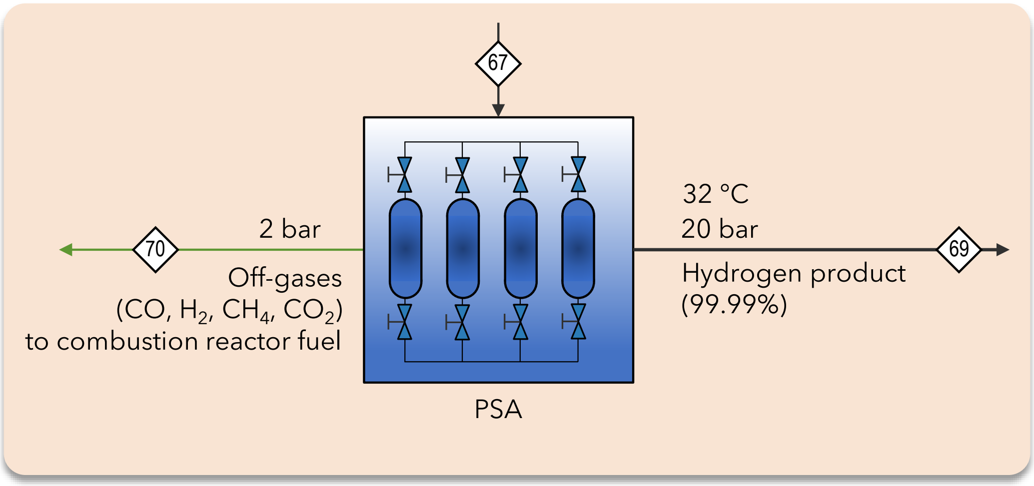

Fig. S24. *PSA PFD for MPW-Hydrogen process.*

Table S27. *Stream summary of MPW-Hydrogen process.* Detailed PFDs for MSW-Methanol process are shown in **Fig. S23 - S24**.

	Stream No. →	63	64	65	66	67	68	69	70
	Temperature, °C	25.0	357.7	344.8	32.0	32.0	32.0	32	23.7
	Pressure, bar	1	20	20	20	20	20	20	2
	Mass Vapor Fraction	0	1	1	0.52	1	0	1	1
	Mass Liquid Fraction	1	0	0	0.48	0	1	0	0
	Mass Solid Fraction	0	0	0	0	0	0	0	0
MT/D	Total Mass flow	816.55	816.55	1472.13	1472.13	762.71	709.41	69.78	692.93
	H_2O	816.55	816.55	837.80	711.74	2.43	709.31	0.00	2.43
	H_2	-	-	78.94	93.04	93.04	-	69.78	23.26
	CO	-	-	217.77	21.78	21.78	-	-	21.78
	CO_2	-	-	336.23	644.16	644.06	0.11	-	644.06
	CH_4	-	-	1.40	1.40	1.40	-	-	1.40

Theoretical maximum hydrogen production from MPW. To better understand the maximum potential of mixed plastic waste to produce hydrogen, a simple stoichiometric targeting analysis is done. Based on the MPW stream elemental composition shown in **Table 2** in the manuscript, the hydrogen content in MPW is 14% by weight. Therefore, the maximum H_2 production solely from MPW without the addition of any external hydrogen is 14 g/100 g waste plastic solid feedstock. The H_2 production can be enhanced by using steam gasification. Assuming CH_2 is the plastic feedstock reactant unit, the steam reforming reaction is:

$-CH_2- + H_2O \rightarrow CO + 2H_2$

Hence, the maximum H_2 yield is 4 g H_2/ 14 g plastic = 28.6 g H_2/100 g MPW.

To further boost the H_2 yield, a water-gas-shift (WGS) reactor can be used. The overall reaction then becomes:

$CO + H_2O \rightarrow CO_2 + H_2$

$\underline{-CH_2- + H_2O \rightarrow CO + 2H_2}$

$-CH_2- + 2H_2O \rightarrow CO_2 + 3H_2$

The maximum H_2 yield for this overall reaction is 6.0 g H_2/ 14 g plastic = 42.9 g H_2/100 g MPW. Hence, the maximum stoichiometric target for hydrogen production from waste plastic feedstock is 42.9 g H_2/100 g MPW.

S6. MSW-Hydrogen process

The gasification and syngas preparation are the same as in the MSW-Methanol process that was described earlier. The water-gas-shift and the PSA sections were described in section **S5** and the PFDs of these sections for the MSW-Hydrogen process are shown in Fig. **S25-S26** respectively. The capital cost for the MSW-Hydrogen process for 240 MT/D MSW processing capacity is $47M with TCI being $93M. The breakdown of capital costs and annual operating costs is shown in **Fig. S5**. The pressure for WGS is 20 bar, not as high as the 80 bar that was needed for methanol synthesis. Hence, the WGS contribution to the CAPEX is a modest 32%. The operating costs are dominated by CO_2 recovery costs, indirect costs that include maintenance labor costs.

Sensitivity analysis on key cost drivers for hydrogen production. The tornado plot for the sensitivity analysis of variables in the MSW-Hydrogen process is shown in **Fig. S8**. The variable with the most impact on hydrogen MSP is the plant capacity. Hence, a larger plant capacity can reduce the hydrogen MSP produced by MSW gasification. Doubling the plant capacity to 500 MT/D MSW feed capacity lowers the hydrogen MSP to $2.62/kg. PSA H_2 recovery is the next critical variable since any hydrogen lost in the PSA is combusted as fuel, thereby reducing H_2 yield. The syngas yield has a modest impact, reducing syngas yield by 30% results in a 17% increase of hydrogen MSP. The base case considers a zero-feedstock price for MSW. Considering an additional income for the MSW-Hydrogen plant from the tipping fee, the hydrogen MSP drops to $2.82/kg, a reduction of 13% from the base case. Since this is a univariate analysis study, a combination of multiple variables changed in the right direction will reduce the hydrogen MSP even further. E.g., for a plant size of 500 MT/D MSW feed capacity, with 90% H_2 recovery in the PSA, the hydrogen MSP drops to $1.89/kg, a reduction of 42% from the base case.

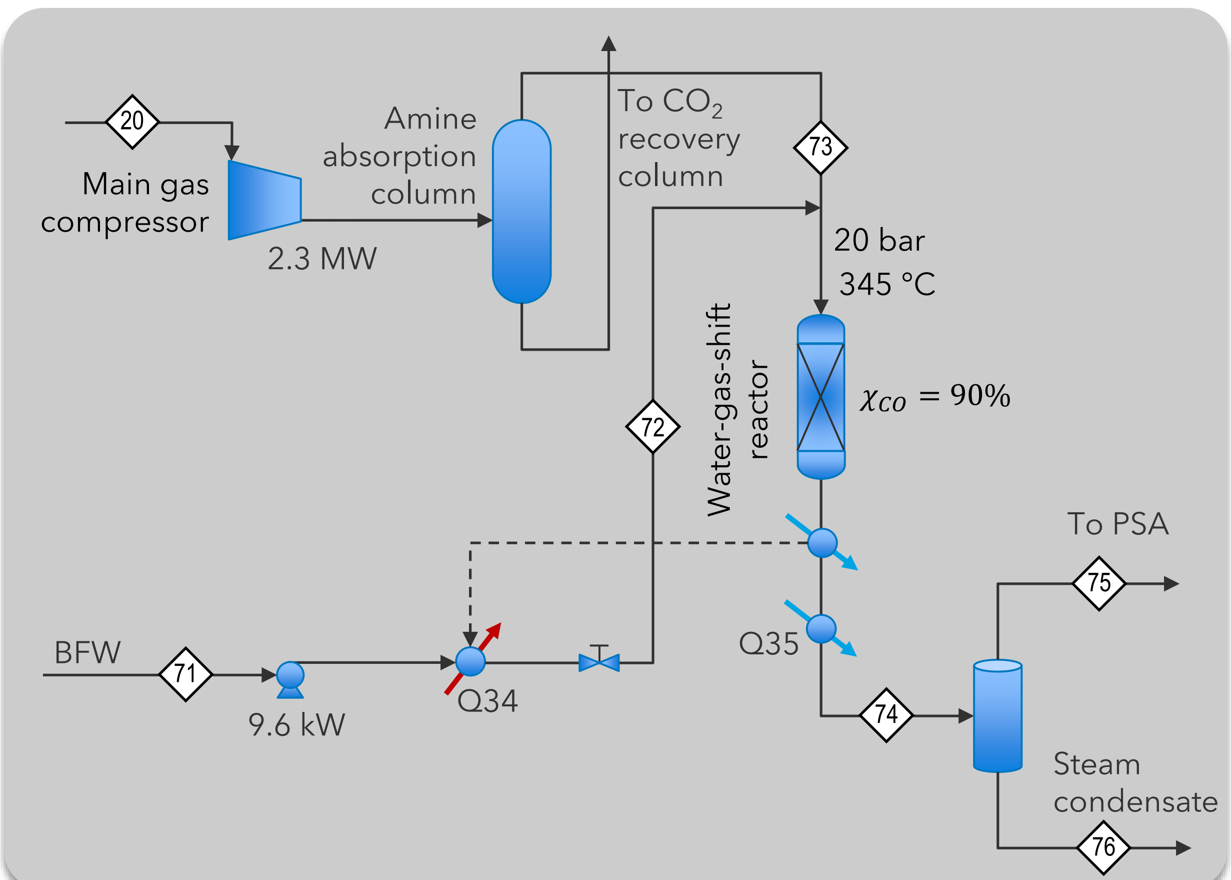

Fig. S25. *WGS PFD for MSW-Hydrogen process.*

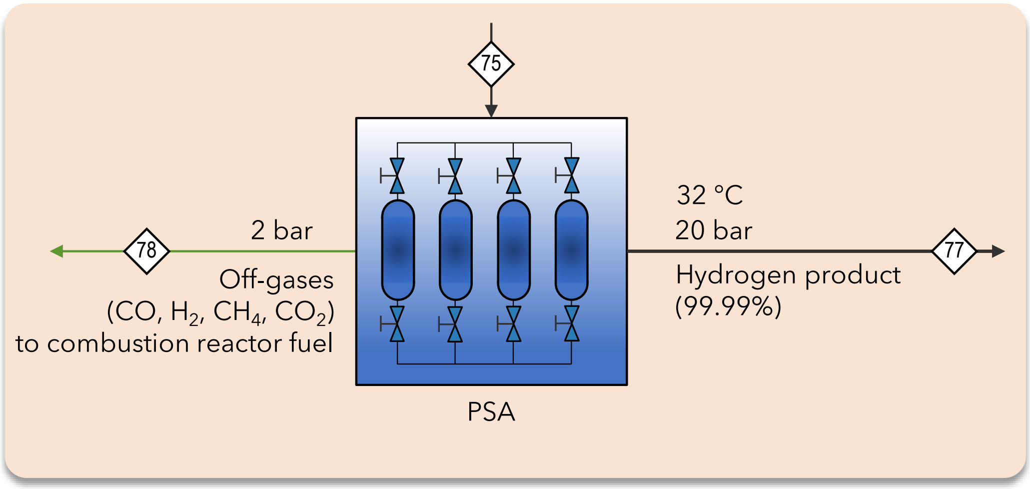

Fig. S26. *PSA PFD for MSW-Hydrogen process.*

Table S28. *Stream summary of MSW-Hydrogen process.* Detailed PFDs for MSW-Methanol process are shown in **Fig. S25-S26**.

	Stream No. →	71	72	73	74	75	76	77	78
	Temperature, °C	25.0	389.3	321.3	32.0	32.0	32.0	32.0	22.0
	Pressure, bar	1	20	20	20	20	20	20	2
	Mass Vapor Fraction	0	1	1	0.58	1	0	1	1
	Mass Liquid Fraction	1	0	0	0.42	0	1	0	0
	Mass Solid Fraction	0	0	0	0	0	0	0	0
MT/D	**Total Mass flow**	151.72	151.72	119.16	270.88	157.83	113.05	22.60	135.23
	H_2O	151.72	151.72	8.08	113.73	0.70	113.04	-	0.70
	H_2	-	-	24.97	30.13	30.13	-	22.60	7.53
	CO	-	-	79.58	7.96	7.96	-	-	7.96
	CO_2	-	-	4.77	117.29	117.28	-	-	117.28
	CH_4	-	-	0.12	0.12	0.12	-	-	0.12
	N2	-	-	1.65	1.65	1.65	-	-	1.65

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