Updates of Hydrogen Production from SMR Process in GREET® 2019

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The hydrogen production pathway using steam methane reforming (SMR) of natural gas (NG) is updated in GREET 2019, based on a recent study by Sun et al. (2019). This study investigated U.S. stand-alone SMR facilities and reported criteria air pollutant (CAP) and greenhouse gas (GHG) emissions per unit of hydrogen production, using SMR facility emission data reported in the National Emissions Inventory (NEI) and the Greenhouse Gas Reporting Program (GHGRP) databases, respectively. The study summarized the CO_2 emission associated with hydrogen production by accounting for emissions both from combustion and chemical conversion processes. The median CO_2 emission normalized for SMR hydrogen production was 9 kg CO_2/kg H_2 production, or 75 g CO_2/MJ H_2 (using H_2 low heating value [LHV]). The median emission is similar with the value of 9.26 kg CO_2/kg H_2 in GREET 2018, which was based on the H2A modeling by Rutkowski et al (2012). For other emissions, the combustion and non-combustion CAP emissions, based on NEI data from Sun et al (2019), reported lower values compared to GREET 2018.

The study by Sun et al (2019) focused on air emissions using the scope of NEI and GHGRP databases, which do not cover energy efficiency. Additional data sources were sought to provide energy use information consistent with CO_2 emission data with reported national median data (i.e., 9 kg CO_2/kg H_2), since energy use and CO_2 emission are directly correlated based on NG feedstock and fuel properties. The energy use data was calculated based on a report by industrial gas supplier Praxair (authored by Bonaquist, 2010), which provided CO_2 emission for each operation step, for a plant producing 100 MMscf H_2 per day (~ 240 metric ton/day). Converting the reported CO_2 emissions to NG energy use by using its EF of 59,399 g CO_2/mmbtu in GREET, we calculated the Praxair SMR energy use (Table 1). The report itemized the CO_2 emissions from different operations steps sourced from energy uses, chemical conversion, i.e., reforming, co-production of steam for export, and power for separation and compression, in addition to NG loss. Overall, about 1.392 mmbtu NG is needed to produce 1 mmbtu H_2 (in LHV). This leads to SMR onsite CO_2 emissions of about 9.4 kg/kg H_2, or 82,658 g_{CO2}/mmbtu H_2, or 78.3 g CO_2/MJ H_2, in LHV, which is close to the median CO_2 from the GHGRP as reported above.

The report by Bonaquist indicates about 6.4 lb steam was exported per lb hydrogen production. The property (temperature and pressure) of the steam was not revealed in the report. Thus, the energy content has to be estimated. The report by Bonaquist indicated that about 290 ton/day CO_2 was emitted from combustion for steam export, which was converted as 161,421 btu energy use per mmbtu hydrogen production (see Table 1). By assuming 90% boiler efficiency, the produced steam is estimated to have energy of approximately 145,000 btu per mmbtu of hydrogen production.

We also considered another scenario in which steam is not exported. In this scenario, the energy used for steam export was omitted. Thus, the overall hydrogen production efficiency via SMR is increased to 81.3 %, and the CO_2 emissions, from both chemical conversion and NG combustion for energy supply, is reduced.

Table 1. CO_2 emission and derived NG energy use for SMR plant with 100 MMscf hydrogen production per day — current practice with steam export (by Bonaquist 2010)

	CO_2	CO_2	CO_2	NG combustion	NG combustion
Unit	ton/day	ton/MMscf H_2	g/mmbtu H_2 (LHV)	mmbtu NG /mmbtu H_2	btu NG/mmbtu H_2
Feed conversion to H_2	1485	14.85	49,099	0.827	826,585
Combustion of fuel for reforming	420	4.2	13,886	0.234	233,782
Combustion for export steam	290	2.9	9,588	0.161	161,421
Power for separation and compression	10	0.1	331	0.006	5,566
External electricity	0	0	0	0	0
NG process fuel	720	7.2	23,805	0.4008	400,768
NG loss	295	2.95	9,754	0.164	164,204
Process fuel (including NG loss)	1,015	10.15	33,559	0.565	564,972
Total actual emission	2500	25	82,658	1.392	1,391,557
Feedstock share in total NG use	=826,585/1,391,557=59.4%				
Process fuel share	= 564,972/1,391,557= 40.6%				
Energy efficiency with steam export	**= 1000000/1,391,557= 71.9%**				

It is worth mentioning that the SMR emissions without steam export were simply derived by subtracting the energy uses and subsequent CO_2 emissions specified for "Combustion for export steam" (in Table 1) from the total emission. However, the actual operation data without steam export might differ in the energy use and emissions from those shown in Table 2, by optimizing the overall process. Thus, we use the SMR process with steam export as the default pathway for hydrogen production from a central plant in GREET.

Table 2. SMR process CO_2 emission and derived NG uses for a plant with 100 MMscf hydrogen production per day — without steam export

	CO_2	CO_2	CO_2	NG combustion	NG combustion
	ton/day	ton/MMscf H_2	g/mmbtu H_2 (LHV)	mmbtu NG /mmbtu H_2	btu NG/mmbtu H_2
Feed conversion to H_2	1485	14.85	49,099	0.827	826,585
Combustion of fuel for reforming	420	4.2	13,886	0.234	233,782
Power for separation and compression	10	0.1	331	0.006	5,566
External electricity	0	0	0	0	0
NG process fuel	430	4.3	14,217	0.2393	239,347.836
NG loss	295	2.95	9,754	0.164	164,204
Process fuel (including NG loss)	725	7.25	23,971	0.404	403,552
Total actual emission	2210	22.1	73,069	1.230	1,230,137
Feedstock share in total NG use	=826,585/1,230,137=67.195%				
Process fuel share	= 403,552/1,230,137= 32.8%				
Energy Efficiency without steam export	**= 1000000/1,230,137= 81.3%**				

The information in Table 1 and Table 2 are incorporated in GREET 2019 to update "user defined" gaseous and liquid H_2 production models. Meanwhile, we also updated the SMR H_2 production pathway based on the H2A model. The key points and changes are summarized below.

1. The H2A model efficiency is unchanged, at 72%. Thus, 1 mmbtu H_2 production consumes 1.388 mmbtu of energy, including 1.371 mmbtu of NG and 0.017 mmbtu of electricity.
2. The NG use share split between feedstock use and process fuel use was updated. By using 0.827 mmbtu NG for SMR feedstock (value from Bonaquist, which is slightly higher than the ideal stoichiometric value of 0.786), the remaining 0.545 mmbtu NG is regarded as process fuel, which also accounts for NG loss. Thus, the updated feedstock NG share is about 59.5%%, whereas in GREET 2018 the feedstock share was 83%.
3. Meanwhile, accounting for both 0.545 mmbtu NG and 0.017 mmbtu electricity as process energy use, the electricity energy share is about 3%.

A comparison of the H2A model values between GREET 2019 and GREET 2018 is shown in Table 3.

Table 3. The comparison of the H2A model values between GREET 2019 and GREET 2018

SMR pathway derived from H2A model	GREET 2019	GREET 2018
Efficiency%	72%	72%
Total NG (mmbtu/mmbtu H_2)	1.371	1.371
Total electricity (mmbtu/mmbtu H_2)	0.017	0.017
NG as feedstock (mmbtu/mmbtu H_2)	1.138	0.827
NG as process fuel (mmbtu/mmbtu H_2)	0.233	0.545
Total input energy (mmbtu/mmbtu H_2)	1.388	1.388
*Feedstock share %	83%	59.5%
Electricity share %	4.4%	3.03%

*In GREET 2018, the feedstock share was calculated as NG feedstock/(total NG); In GREET 2019, the feedstock share was re-defined and calculated as NG feedstock/(total NG + electricity);

For the SMR process, whether based on the Praxair report by Bonaquist, or the H2A model by Rutkowski, the CAP emission (VOC, CO, NO_x, SO_x, PM_{10}, and $PM_{2.5}$) was updated, based on the study by Sun et al. (2019). For CAP emissions, the Sun et al. study showed lower combustion emissions relative to previous data in GREET 2018, as shown in Table 4.

Table 4. SMR process onsite CAP emissions (g/mmbtu H_2)

CAP pollutant	GREET 2019/study by Sun et al. (2019)		GREET 2018	
	Combustion EF	Non-combustion EF	Combustion EF	Non-combustion EF
VOC	1.249	0.965	0.591	0.760
CO	1.239	1.261	5.169	1.236
NO_x	5.731	1.645	8.472	2.304
PM_{10}	1.724	1.207	0.816	1.035
$PM_{2.5}$	1.724	1.164	0.816	1.009
SO_x	0.054	0.027	0.063	0.159

The non-combustion CAP emission data (per mmbtu of H_2 production) in Table 4 are incorporated in the hydrogen tab in GREET 2019.

The NG combustion-related CAP emissions for SMR were estimated by converting the hydrogen-based emission values to NG-based emissions using the 0.565 mmbtu share of NG combusted per 1 mmbtu H_2; see Table 1. The derived NG combustion EF for SMR process are shown in Table 5, which are added as new data in the EF tab in GREET 2019. These values are also compared to NG combustion EFs for other combustion technologies (see Table 5). The values of BC (black carbon) and OC (organic carbon) were not reported by Sun et al. (2019); thus comparable data from Utility/Industrial Boiler (>100 mmBtu/hr input) are used as surrogates for BC and OC.

Although CH_4 and N_2O data are reported by Sun et al. (2019), we applied data from Utility/Industrial Boiler (>100 mmBtu/hr input) for SMR in GREET 2019. This is because fewer facilities reported CH_4 and N_2O data in the GHGRP database for SMR plants, compared to the much larger number of facilities reporting CAP emissions in the NEI database.

Table 5. SMR combustion CAP emissions compared to industrial NG use in different combustion technologies (g/mmbtu NG)

	GREET 2019	GREET 2018 (unchanged for 2019 version)			
CAP pollutant	SMR NG	Utility/Industrial Boiler (>100 mmbtu/hr input)	Small Industrial Boiler (10-100 mmbtu/hr input)	Large Gas Turbine	CC Gas Turbine
VOC	2.211	2.540	2.540	1.056	0.267
CO	2.194	22.210	24.970	41.286	14.533
NO_x	10.145	36.400	41.050	31.969	17.425
PM_{10}	3.051	3.507	3.507	3.575	0.133
$PM_{2.5}$	3.051	3.507	3.507	3.575	0.133
SO_x	0.095	0.269	0.269	0.269	0.269
BC	0.579	0.579	0.579	0.104	0.004
OC	1.501	1.501	1.501	2.431	0.090
CH_4	1.060	1.060	1.060	1.056	1.142
N_2O	0.750	0.750	0.350	0.102	0.119
CO_2	59,399	59,367	59,363	59,342	59,386

All NG combustion has similar CO_2 emission, given the CO_2 emissions are directly linked to fuel carbon content, with minor adjustment for the carbon content in CH_4, VOC, and CO. We note that the NG combustion for SMR has similar CAP EF compared to most industrial NG combustion technologies.

References

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