Catalytic Wet Gasification of Municipal and Animal Wastes

Kyoung S. Ro,*,† Keri Cantrell,† Douglas Elliott,‡ and Patrick G. Hunt†

USDA-ARS Coastal Plains Soil, Water & Plant Research Center, 2611 W. Lucas Street,
Florence, South Carolina 29501-11241, and Pacific Northwest National Laboratory, P.O. Box 999,
MSIN K2-12, Richland, Washington 99352

Currently there is worldwide interest in deriving energy from bio-based materials via gasification. Our objective was to assess the feasibility of wet gasification for treatment/energy conversion of both animal and municipal wastes. Wet wastes such as swine manure and raw sewage sludge could be processed directly via current wet gasification technology. Furthermore, these wastes generated high amounts of net energy based on reaction material and energy balances. Without use of an efficient heat recovery system, municipal solid wastes and unpaved feedlot manure would not generate positive energy return from wet gasification. Due to high sulfur content of the wastes, pretreatment to prevent the poisoning of catalysts is essential. The costs of a conceptual first generation wet gasification manure management system for a model swine farm were significantly higher than that of the anaerobic lagoon system. However, there are many significant environmental advantages of the wet gasification, e.g., BOD removal, odor elimination, and pathogen kill.

Introduction

Traditionally nutrients in animal manure have been recycled by applying manure to cropland and pastureland to promote plant growth. However, over the last few decades, confined animal feeding operations (CAFOs) in the United States and many other countries have undergone extensive expansions and consolidations.[1] This shift of animal agriculture toward fewer, but larger, operations has created environmental concerns in recycling and disposal of animal manure.[2] The production of animal manures often exceeds assimilative capacity of local land. Excess land application of animal manure leads to both air and water quality problems.[3] These problems frequently involve nitrogen, phosphorus, trace metals, ammonia, odors, and pathogens. In addition, estrogens and pharmaceutically active compounds (PACs) in runoff may harm the ecosystems and the environment.[4,5] The problems are often exhibited in deteriorated stream, river, and estuarine quality. Thus, surplus manure must be hauled to other sites with adequate land for application and assimilation of manures. If other land is not available, the manure must be processed to maintain environmental integrity.

The common means of processing animal manures have been composting and anaerobic digestion. Composting aerobically processes manure and organic materials via microbial assimilation and heat production. The heat could be managed to develop mesophilic temperatures (about 35 °C) to thermopilic temperatures (about 55 °C).[6] Composted manure is a relatively odorless, pathogen-free substance containing plant nutrients, which allows its use as soil amendments. Unfortunately, composting manure requires manure dewatering, longer process times of several months, and potential emission problems.[7,8]

Anaerobic digestion degrades and stabilizes the manure's organic material via two-stage biological processes. The first stage involves the breakdown of complex organic matter into less-complex carbon compounds; this step is followed by methanogenic bacteria converting carbon compounds into gases, mainly methane and carbon dioxide. Trace gases such as ammonia, oxygen, and hydrogen sulfide can also be found in the biogas. Anaerobic lagoons promote anaerobic digestion process in open lagoons. These lagoons are commonly used by CAFOs to treat and store liquid swine manure and allow the product biogas to escape into the atmosphere. The escaped biogas may become the source of air pollution.[9,10] In contrast to lagoons, anaerobic digesters use a gas-tight tank or impermeable cover placed over a lagoon to capture the biogas. The captured biogas is typically used as a space heating fuel. Although these technologies reduce pathogens and produce a more uniform, high-quality fertilizer, proper disposal or land application of the solids is still required. In addition, as with composting process, these technologies require long process time and large facilities due to slow anaerobic processes involved.

Unlike the above conventional technologies based on physicochemical and biological methods, thermochemical processes such as gasification technologies may be used not only to convert animal manure into environmentally acceptable forms, but to also harness the chemical energy stored in animal manure and produce energy-value-added products. In fact, animal manure represents a significant source of renewable bioenergy; currently animal manure provides 35 million dry tons of sustainable biomass feedstock per year which is 18% of the entire sustainable biomass feedstock from the U.S. agriculture lands.[11] Among various thermochemical technologies such as pyrolysis and various gasification technologies, wet gasification or hydrothermal gasification offers the unique advantage of directly using wet manure feedstocks, thus eliminating the need for dewatering and drying pretreatments.

Within a single step, wet gasification converts organic matter into methane and carbon dioxide and destroys harmful organics. Other positive aspects of wet gasification include no fugitive emissions to the atmosphere and no residual organics needing disposal. Whereas oxygen is not required (water reacts with the organics), the process is simpler and less costly than other aerobic thermochemical processes such as wet-air oxidation or supercritical oxidation processes. It is much faster than the composting or anaerobic digestion methods; the residence time is roughly 15 min compared to weeks and months for biological treatment processes. Furthermore, there is no inherent formation of additional waste such as the digested sludge. These advan-

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* To whom correspondence should be addressed. Tel.: 843-669-5203 x107. Fax: 843-669-6970. E-mail: kyoung@florence.ars.usda.gov.

† USDA-ARS Coastal Plains Soil, Water & Plant Research Center.

‡ Pacific Northwest National Laboratory.

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Table 1. Wet Gasification Reactions of Various Animal and Municipal Wastes

feedstock wastes	wet gasification reaction	reaction number
wet manure (TS[a] < 15%)		
swine manure[23] ($CH_{1.69}N_{0.06}O_{0.51}$)	$CH_{1.69}N_{0.06}O_{0.51}(s) + 0.39H_2O(l) \rightarrow 0.55CH_4(g) + 0.45CO_2(g) + 0.063NH_3(aq) + 0.01H_2(g)$	(1)
dairy manure[24] ($CH_{1.57}N_{0.04}O_{0.64}$)	$CH_{1.57}N_{0.04}O_{0.64}(s) + 0.32H_2O(l) \rightarrow 0.52CH_4(g) + 0.48CO_2(g) + 0.04NH_3(aq) + 0.01H_2(g)$	(2)
dry manure (TS > 15%)		
poultry litter[25] ($CH_{1.45}N_{0.07}O_{0.56}$)	$CH_{1.45}N_{0.07}O_{0.56}(s) + 0.12H_2O(l) \rightarrow 0.48CH_4(g) + 0.52CO_2(g) + 0.07NH_3(aq) + 0.08H_2(g)$	(3)
unpaved feedlot manure[26] ($CH_{1.45}N_{0.08}O_{0.5}$)	$CH_{1.45}N_{0.08}O_{0.5}(g) + 0.48H_2O(l) \rightarrow 0.51CH_4(g) + 0.49CO_2(g) + 0.08NH_3(aq) + 0.07H_2(g)$	(4)
paved feedlot manure[26] ($CH_{1.45}N_{0.06}O_{0.47}$)	$CH_{1.45}N_{0.06}O_{0.47}(s) + 0.59H_2O(l) \rightarrow 0.47CH_4(g) + 0.53CO_2(g) + 0.06NH_3(aq) + 0.28H_2(g)$	(5)
municipal wastes		
raw sludge[27] ($CH_{2.07}N_{0.06}O_{0.57}$)	$CH_{2.07}N_{0.06}O_{0.57}(s) + 0.43H_2O(l) \rightarrow 0.5CH_4(g) + 0.5CO_2(g) + 0.06NH_3(aq) + 0.37H_2(g)$	(6)
MSW[28] ($CH_{1.56}N_{0.03}O_{0.55}$)	$CH_{1.56}N_{0.03}O_{0.55}(s) + 0.39H_2O(l) \rightarrow 0.53CH_4(g) + 0.47CO_2(g) + 0.03NH_3(aq) + 0.07H_2(g)$	(7)

[a] TS = total solid content (% m/m).

tages make wet gasification a potentially effective alternative manure management technology. However, it has not been extensively evaluated to date for managing animal manures.

The objectives of this paper are to (1) further introduce wet gasification technology, (2) conduct feasibility analyses of wet gasifying various animal and municipal wastes by analyzing energy production potentials and end product compositions, and (3) compare relevant costs and benefits of first-generation wet gasification technology to anaerobic lagoon animal manure management technology.

Wet Gasification Technology

The U.S. Department of Energy (DOE) and the Gas Research Institute supported the original development of the wet gasification technology for treatment of organic residuals. The wet gasification process is capable of treating a broad range of different organic chemical functional types containing carbon, hydrogen, and oxygen.[12,13] It uses catalytic hydrothermal processing conditions (250−360 °C, up to 22 MPa) to treat wet biomass, organics-in-water process residues, and wastewater. It converts the organic contaminants to gases composed of mainly methane and carbon dioxide. Through a metal catalyst, gasification of wet biomass can convert high levels of carbon to gas at a relatively low temperature (350 °C).

Developing the metal catalyst for wet gasification has been an important factor in making this technology viable. Previous reports of continuous reactor experiments with biomass feedstocks provided preliminary short-terms processing results[14,15] but also showed the problems of long-term operation of the process with contaminants inherent in biomass. Attempts to pretreat biomass by removing certain components, like alkaline earths, to allow extended use with catalysts, have also been documented.[16] In recent publications, Elliott and co-workers[17,18,19] reported more stable catalyst formulations for wet gasification based with ruthenium. In a pressurized-water environment (20 MPa), near-complete conversion of the organic structure of biomass to gases has been achieved in the presence of a ruthenium metal catalyst. The process is essentially steam reforming; there is no added oxidizer or reagent other than water. The gas produced is a medium-heating value gas containing high levels of methane, as dictated by thermodynamic equilibrium.[20]

Elliott et al.[21] also examined the use of the wet gasification technology for agricultural residuals such as dairy manure and distiller's dried grains and solubles. The organic material in these wastes was converted to a gas containing primarily methane and carbon dioxide leaving an effluent with a COD usually below 1000 mg/L. This reaction was conducted in several bench-scale reactor types including a batch reactor and a continuous-flow stirred-tank reactor, allowing them to obtain kinetic data for the process. In developing the engineering concepts behind wet gasification, Elliott et al. are currently operating two continuous-flow tubular reactor systems: a bench scale system (1−2 L/h) and a scaled-up reactor system (5 gal/h). Results with organic wastes in these systems were quite encouraging; high conversions of organic material to gas were achieved in <20 min at 350 °C in the plug−flow, tubular reactor. While good gas production was demonstrated, biomass trace components caused some processing difficulties in the fixed catalyst bed tubular reactor system. Inorganic components are, for the most part, unaffected in the process. Yet they may act as poisons at high concentrations, and feedstocks may require pretreatment. For example, nitrate and cyanide are destroyed in the process producing nitrogen gas and ammonia; however, sulfides and sulfates are catalyst poisons and must be avoided or removed.

On the basis of the process knowledge gained from the previous wet gasification experiments, the following sections evaluate the energetics and applicability of the wet gasification technology for treating various animal manures, sewage sludge, and municipal solid wastes (MSW).

Wet Gasification Reactions of Animal and Municipal Wastes

Chemical empirical formulas for various animal manures, sewage sludge, and MSW were estimated based on the elemental compositions. These formulas were used to balance the wet gasification reactions as shown in Table 1. To evaluate the adequacy of these reactions, product gas compositions of previous wet gasification experiments were compared.[19] Equation 2 (see Table 1) predicted the product gas composition of the dairy manure wet gasification experiment reasonably well. The bench-scale wet gasification of dairy manure produced 54% CH_4, 45% CO_2, and 1% H_2, while reaction 2 predicted 51% CH_4, 48% CO_2, and 1% H_2. For all waste materials examined, about half of the carbon was converted to CH_4 and the other half to CO_2. All nitrogen was assumed to convert to aqueous NH_3.[13] Very little hydrogen was produced from any of the wet gasification reactions in Table 1. Wet gasifying animal manures

Table 2. Action Energies for Wet Gasification of Animal Manures and Municipal Wastes

compound	$\Delta H_{c,298}$ (kJ/mol)	swine manure	dairy manure	poultry litter	unpaved feedlot manure	paved feedlot manure	raw sewage sludge	MSW
$CH_4(g)$	−890.4	−490	−463	−427	−454	−419	−445	−472
$CO_2(g)$	0	0	0	0	0	0	0	0
$NH_3(aq)$	−348.5[a]	−22	−14	−25	−26	−22	−22	−9
$H_2(g)$	−285.8	−11	−3	−23	−20	−80	−106	−20
$H_2O(l)$	0	0	0	0	0	0	0	0
HHV of feedstocks (kJ/mol)[b]		460[29]	504[29]	453[29]	429[29]	461[26]	375[30]	507[31]
ΔH_{rxn} (kJ/mol)[c]		−63	25	−22	−71	−60	−198	6
energy value of product gases (kJ/mol)		501	466	450	474	499	551	492
ash content (%)		12.7[23]	17[29]	20[29]	58.7[26]	20.2[26]	35[28]	48[31]
energy value of product gases (kJ/kg DM)		19156	15863	13832	8666	18170	14933	11257

[a] Estimated from $4NH_3(g) + 3O_2(g) \rightarrow 2N_2(g) + 6H_2O(l)$. [b] High heat value (HHV) of volatile fraction of dry matter characterized with the empirical formula. [c] Heat of reaction.

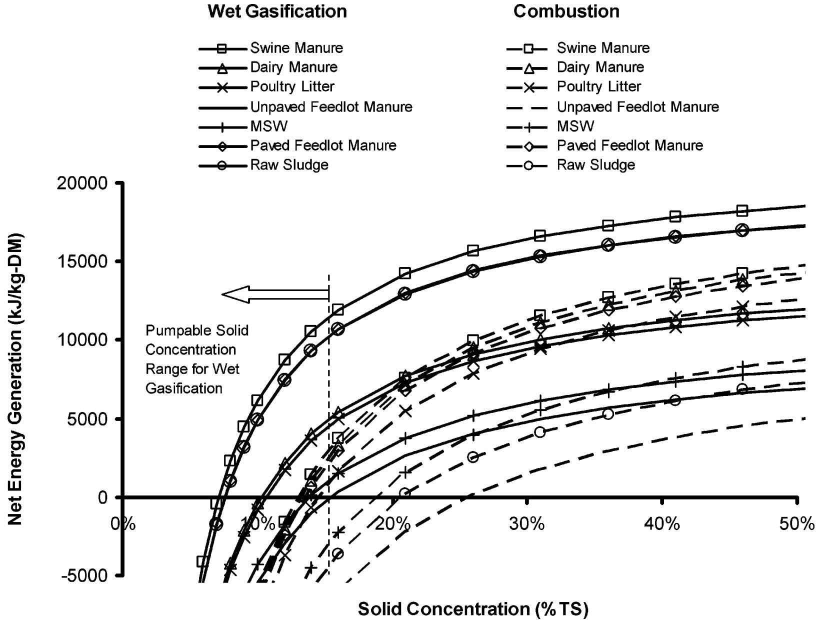

Figure 1. Net energy production from wet gasifying various waste materials.

would produce significant amounts of ammonia. Based on the USDA manure production and fertilizer consumption statistics,[22] the ammonia-N produced from wet gasifying swine, dairy manures, and poultry litter would account for 10% of the entire 2005 U.S. consumption of 12.3 × 10^6 tons of nitrogen for plant nutrition.

Wet gasification reaction energies of these wastes were also estimated using the heat of combustion ($\Delta H_{c,298}$) of the components as shown in Table 2. The wet gasification reactions were more-or-less thermally neutral for most waste materials except for the raw sludge which was significantly exothermic. While wet gasification reactions of swine manure, poultry litter, and feedlot manures were slightly exothermic, those of dairy manure and MSW were slightly endothermic. Wet gasifying raw sewage sludge produced product gases (energy mostly CH_4 and H_2) with the highest energy per mole; however, swine manure wet gasification produced product gases with the highest energy per kilogram of dry matter (DM) due to its lower ash content (12.7% vs 35%). The product gases from wet gasifying unpaved feedlot manure would contain the lowest energy value due to very high ash content of the manure (58.7%).

Portions of the product-gas energy could be used to bring the feedstock materials to the wet gasification operating conditions (350 °C and 20 MPa) and supply additional energy if the reaction is endothermic. Because feedstock wastes were mostly water, enthalpies of water were used to estimate the shaft work (E_{W_s}) necessary to bring the feedstock to the wet gasification operating conditions. The net energy generated from wet gasifying 1 kg of dry waste was estimated as

$$E_{total} = E_{CH_4+H_2} - E_{W_s} + E_{rxn} \tag{8}$$

where E_{total} = net energy generated from wet gasifying 1 kg of dry matter (given in kilojoules); $E_{CH_4+H_2}$ = energy value of CH_4 and H_2 (given in kilojoules); E_{W_s} = shaft work necessary to bring feedstock to 350 °C and 20 MPa (given in kilojoules); and E_{rxn} = reaction energy (given in kilojoules; positive for exothermic and negative for endothermic reactions).

The net energy produced per kilogram of dry waste increased with solid contents as shown in Figure 1. Thermal energies produced from combusting these wastes were also shown in Figure 1. Wet gasification generally generated more energy than combustion especially at lower solid contents. Wet gasification of swine manure generated the highest positive net energy followed by raw sludge and paved feedlot manure, generating virtually the same energy. The threshold solid concentration for these wastes was about 8%. The threshold solid concentration is an energy break even point, above which the process is a net energy generator, when considering all process energy requirements, i.e., pumping and heat loss. Poultry litter and dairy manure generated approximately half of the swine manure. Unpaved feedlot manure and MSW with high ash contents

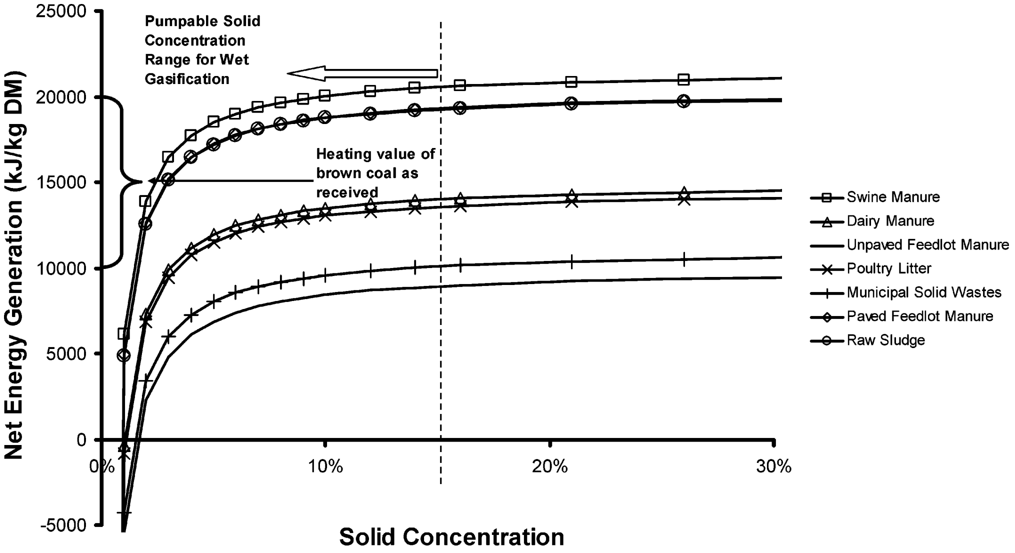

Figure 2. Net energy production from wet gasification with the 90% efficient heat recovery system.

generated the lowest energy with a threshold solid concentration of about 15%. Because current wet gasification technology can handle only a liquid form of feedstocks (i.e., moisture contents higher than 85% or solid content less than 15%), unpaved feedlot manure could not be processed without spending extra energy. This limitation arises from the wet gasification processing requirement that feedstock must be pumpable as a liquid. Wet feedstocks such as swine, dairy manures, and raw sludge with typical moisture contents of about 85−95% could be fed directly into the wet gasification system. For dry feedstocks such as poultry litter and MSW with a high solid concentration of about 79%, a substantial amount of water must be added before it could be wet gasified. Instead of adding extra water, wet and dry feedstocks could be mixed to provide the necessary moisture to make the mixture pumpable. For instance, a mixture of 93 kg of fresh swine manure (90% moisture) and 7 kg of poultry litter (25.5% moisture) would have a moisture content of about 85.2%. This mixture could then be wet gasified. However, this option is not always feasible because it depends on the proximity of the feedstock sources.

The threshold solid concentration would decrease if a significant portion of product gas stream heat could be recycled to heat the incoming feedstock. In fact, Elliott et al.[32] developed a double-tube heat exchanger that could recycle up to 90% of the energy to raise the feedstock temperature. Assuming that wet gasification system is equipped with the 90% heat recovery system, all feedstock wastes would generate net positive energy at solid concentrations greater than 2% as shown in Figure 2. One should note that wet gasifying swine manure produced comparable energy derived from combusting brown coal (Figure 2). Because of its high-energy production potential and its ideal moisture contents for wet gasification, swine manure was selected for further economical analyses based on a conceptual, modular wet gasification system for a model swine farm.

Model Farm Wet Gasification System

The application of wet gasification technology to swine manure has not been experimentally tested, much less commercialized. The subsequent analyses were conducted based on a conceptual wet gasification system for a 4400-head, feeder-to-finishing model swine farm. It was assumed that the model farm would produce about 1580 L/h of flushed swine manure from the animal houses. Figure 3 shows the flow diagram of the proposed wet gasification swine manure management system for the model swine farm. The flushed swine manure is directly treated with wet gasification system. The product gases mostly of CH_4 and CO_2 can be readily used as space heating or further processed thermocatalytically to produce liquid fuels. The ammonia in the product water stream can be recovered via stripping or membrane separation. The rest of water can be recycled back to the swine farm as a drinking water for pigs after minimal treatment. The costs-and-benefits analysis of both traditional anaerobic lagoon and the wet gasification systems for this model swine farm was conducted and compared.

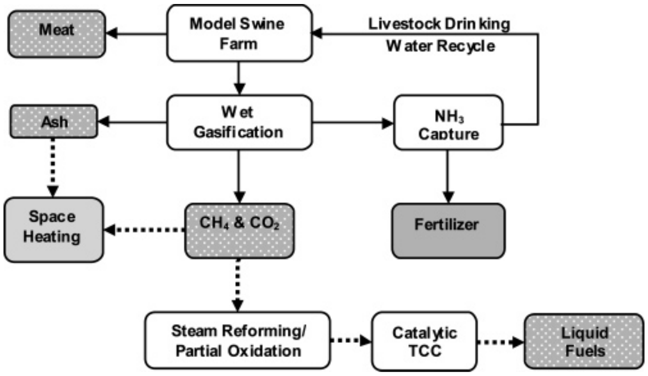

Figure 3. Wet gasification swine manure management system.

A detailed process flow diagram of the farm-scale wet gasification unit is depicted in Figure 4. The major equipment pieces were designed and costed based on 2 LHSV (liquid hourly space velocity; this is the volume of manure per volume of catalyst bed per hour) and a simple spreadsheet calculator developed for the technology at the Pacific Northwest National Laboratory. In addition, considering the swine manure composition, a means must be found to handle the mineral and sulfur content in the feedstock to prevent it from poisoning the catalyst and plugging the bed. The process concept included important modifications that are currently under development to allow processing of biomass feedstocks containing mineral matter and reduced sulfur (generally as protein).

Net Energy from Model Swine Farm Wet Gasification

The net energy generated from wet gasifying flushed swine manure of various solid contents was estimated according to

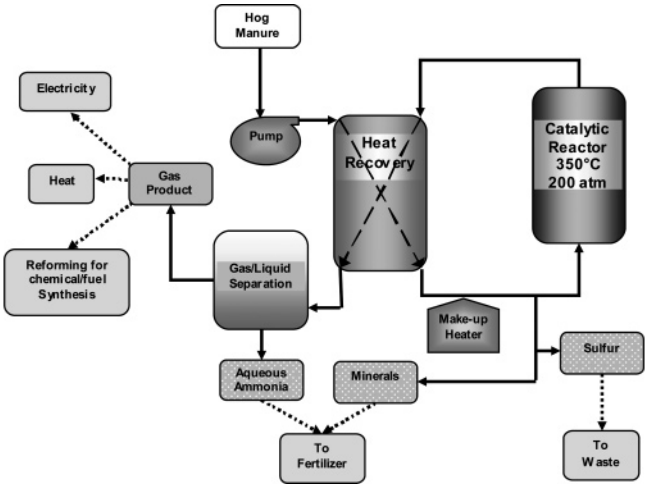

Figure 4. Process flow for the farm-scale, conceptual wet gasification unit.

eq 8. As the solid content of feedstock swine manure increased, the net energy from gasification also increased as shown in Figure 5. The threshold solid content was about 8% above which the wet gasification process generated positive net energy output (see also Figure 1). Simple swine manure combustion yielded much less net energy, and the threshold solid level was higher, i.e., about 12%. However, the use of the 90% efficient heat recovery system greatly reduced the threshold solid concentration to about 1%.

Economics of Wet Gasification System

The capital and operating costs of the conceptual wet gasification system are as shown in Table 3. The installed equipment cost included the estimates of the equipment cost and uses a factor method to determine installation costs as well as piping, insulation, and instrumentation costs. The major equipment items included the high-pressure pumps (including a back-up pump), a heat exchanger for heat recovery for preheating the feedstock, a fired heater for heat-loss makeup, mineral and sulfur separators, fixed-bed catalytic reactors, product separators, and feed and product tanks. The installed capital cost of the farm-scale wet gasification unit for the 4400-head swine farm was $991 000. The annual operating cost was $218 359. As for the positive cash flow, the wet gasification would produce valuable product gases worth $47 006/yr.

Table 3. Cost Data for Wet Gasification of Swine Manure

		operating costs	annual cost
installed capital cost	$991 000	depreciation	$49 534
credit for gas product (in addition to meeting process requirement)	$47 006/year	catalyst[a]	$43 762
		sulfur scrub	$3 198
		utilities	$2 721
		labor	$54 750
		maintenance	$49 534
		taxes/insurance	$14 860
		total operating costs	$218 359

[a] A credit of $39 018/year for catalyst recovery has been considered for the catalyst operating cost.

The operating costs included catalyst makeup cost, which depends on (1) catalyst lifetime (assumed here at 2 years) and catalyst recovery credit for the spent catalyst (credit was assumed in Table 3); (2) labor (priced at $25/h) and without supervisory labor cost as might be expected in farm setting; and (3) electrical utility costs at 5.5 cents per kWh.[33] The sulfur scrubbing component was assumed to be regenerable and not directly disposed material. However, the extent and means of regeneration has not been demonstrated (100 cycles was assumed here before disposal). The means of regeneration could be extrapolated from other applications, such as dry box cleaning, in which nickel materials were regenerated by carefully controlled methods with more than 100 cycles. The fate of the sulfur would be as a permitted atmospheric release of SO_x from an incinerator because a small-scale elemental sulfur recovery system would not be economical. Maintenance, taxes, and insurance were costed by a factor method based on the capital cost; the capital cost was applied by straight-line depreciation with a lifetime of 20 years.

Annualized costs of the wet gasification system were compared with that of the conventional anaerobic lagoon system. Using 8% interest rate, the annualized costs including both capital and operating costs for the wet gasification system is $375 per animal unit (AU, 1000 lb live weight). The capital and operating costs of the wet gasification system is significantly higher than that for the anaerobic lagoon system (about $85−$95 annualized cost per AU[34]). However, considering the fact that the anaerobic lagoon system is already established technol-

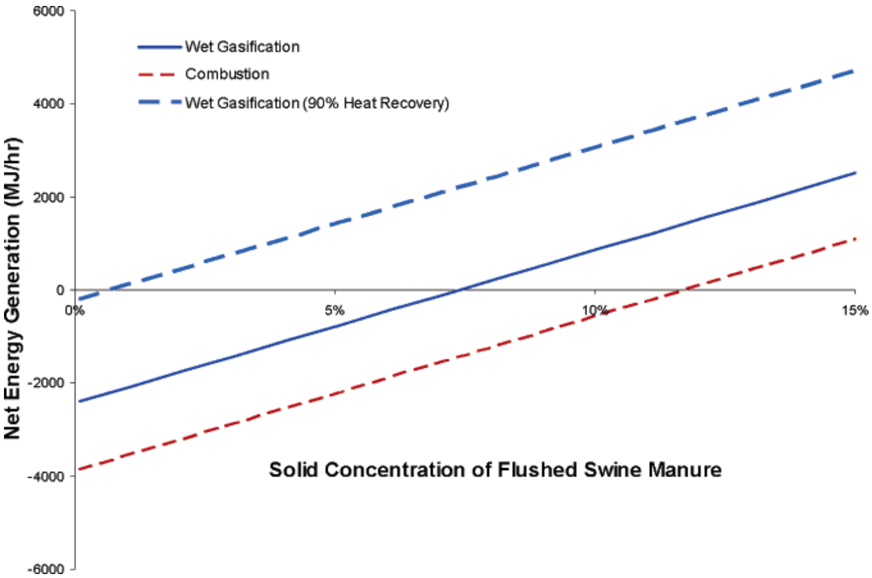

Figure 5. Net energy productions from wet gasifying and combusting swine manure of the model swine farm.

ogy while the wet gasification system is still in the research and development stage, the costs of the wet gasification system will decrease as the technology matures. In addition, our initial design value of 2 LHSV may be increased to 4 LHSV through additional catalyst research, which could substantially lower the capital costs.

Conclusions and Recommendations

Process Applicability. Wet feedstock wastes such as swine, dairy manures, and raw sewage sludge with high moisture contents can be directly gasified to produce an energy rich product gas; this gas can be used directly for space heating or further processed to produce biofuels through steam reforming and other catalytic processes. However, additional water must be added to dry feedstock wastes such as poultry litter, MSW, and feedlot manures to make the feedstock pumpable. Without any heat recovery system, wet gasifying MSW and unpaved feedlot manure would not produce positive energy return due to high ash contents of these materials. With an efficient heat recovery system (90% efficiency), all feedstock materials produced net positive energy return at solid contents higher than 2%. Wet gasification of swine manure produced the highest number of net energy; MSW and unpaved feedlot manure produced the lowest. With the 90% efficient heat recovery system, even the lowest energy producing feedstock such as unpaved feedlot manure produced about half of the energy from combusting brown coal (8900 kJ/kg feedlot manure vs average 15000 kJ/kg brown coal).

Process Costs. Capital and operating costs for the wet gasification system for a model swine farm were higher compared to that of the conventional anaerobic lagoon system. However, the high rate of conversion of the organic matter into gas drastically decreases the land requirement for manure application, which also leads to reductions in transportation and fuels charges and tipping fees. Wet gasifying swine manure offers additional benefits to farmers such as the following:

(i) It destroys pathogens, most active organic compounds such as estrogens, antibiotics, and odorous compounds (all these are a nuisances and harmful to humans, livestock animals, and the environment);

(ii) It produces relatively clean water, which could be used as drinking water for livestock animals with a minimal treatment (this could lead to an additional reduction in potable water usage translating into reduced overall utility costs for the farm); and

(iii) It produces valuable byproducts such as ammonia and phosphates which have potential on the fertilizer market.

Because further testing and validation of the overall impact wet gasification on a farm needs to be done, potential monetary savings of the above benefits are not included in the economical analysis. These added environmental benefits could substantially reduce the actual costs of the wet gasification technology and overall impact swine farms have on the surrounding ecosystem.

Further Improvements Needed To Prevent Poisoning of Catalyst. The removal of feedstock contaminants is a prerequisite for catalytic driven gasification processes. These contaminants include precipitating minerals like calcium, magnesium, and phosphorus, as well as sulfur, which poisons the metal catalyst. Due to the high level of sulfur in the animal manures and raw sewage sludge, sulfur removal by reactive absorbent must be accompanied with a simple regeneration process for the adsorbent. Development of contaminant removal steps and their demonstration will be required before application of catalytic hydrothermal gasification as a swine manure treatment.

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