

Integrated hydrogen energy systems for power and transportation

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The purpose of this research is to develop biomass-to-hydrogen systems that are suitable for fuel cell-based power and transportation. Our approach is thermal gasification of biomass followed by a biological gas-conditioning step to increase the yield of hydrogen. Municipal solid waste, sewage sludge, agricultural (manure) and forestry wastes (plant byproducts, wood), are chemically heterogeneous substrates that are only slowly metabolized by a complex community of microorganisms, which convert the organic matter, ultimately, to carbon dioxide (CO₂) and methane (CH₄). Alternatively, dried, low-grade organic matter can be thermally gasified to produce a fairly homogeneous mixture of gases comprised, primarily, of carbon monoxide (CO) and hydrogen (H₂), with smaller amounts of carbon dioxide (CO₂), hydrogen sulfide (H₂S), and CH₄.

Gasification usually consists of two distinct processes: combustion and pyrolysis. Pyrolysis is the chemical decomposition of solid fuel at elevated temperatures to produce a combustible gas mixture. Since pyrolysis is an endothermic reaction, it must be accompanied by a heat source. Conventional gasification admits sufficient air or oxygen to the reactor to burn part of the fuel, thus releasing heat to support pyrolysis of the rest of the fuel. Gas produced in air-blown gasifiers typically have heating values only 10 to 20% that of natural gas. This low heating value is largely the result of nitrogen diluting the fuel gas. Oxygen could be used as the gasification agent, but high capital costs preclude this from consideration at the relatively small sizes envisioned for most biomass energy systems.

An alternative approach, known as indirectly heated gasification, physically separates combustion and pyrolysis. As a result, the products of combustion do not appear in the fuel gas. Higher heating values of 14.2 to 16.6 MJ Nm⁻³ (382 to 445 Btu scf⁻¹) on a dry basis have been demonstrated. Several schemes have been suggested for transporting heat from the combustion reactor to the pyrolysis reactor. These include transferring hot solids from the combustor to the pyrolyzer, transferring a chemically regenerative heat carrier between two reactors, and transferring heat through a wall common to the reactors. These schemes are not well suited to the relatively small scales envisioned for rural heat and power generation.

An indirectly heated ballasted gasifier that uses a single reactor for both combustion and pyrolysis has been developed. Instead of spatially separating these processes, they are temporally isolated. The producer gas is not diluted with the products of combustion or the nitrogen from air. Heat released during combustion is stored as latent heat in phase change material sealed in tubes immersed in the reactor. This heat is released during the pyrolysis stage of the cycle. The phase change material may be an inorganic salt or metal alloy. The reactor employs a fluidized bed to obtain uniform and rapid distribution of heat from the phase change material to the pyrolyzing fuel. The ballasted gasification system converts biomass into a "producer" gas mixture consisting of approximately 40% CO, 25% H₂, 18% CO₂, 10% CH₄, plus smaller fractions of other hydrocarbons (C₂H₂- 0.5%, C₂H₄- 5.2%, C₂H₆- 0.5%). These hydrocarbons can be catalytically steam reformed to additional hydrogen and carbon monoxide, yielding in excess of 50% hydrogen in the producer gas.

For transportation applications, the producer gas is cooled and processed through a biologically based gas conditioning system, subsequently described, which converts CO and water into hydrogen and carbon dioxide. The final gas mixture is approximately 58% hydrogen and 18% carbon dioxide. The hydrogen is separated and compressed to high pressure for transportation applications, which typically employ proton exchange fuel cells (PEMFC).

For electric power applications, the hot, steam-reformed producer gas is fed directly to a solid oxide fuel cell (SOFC). Pyrolysis of biomass at a flow rate of approximately 45g/s will generate approximately 2.6 moles/s H₂, an amount sufficient to fuel a 250 kW SOFC (the heat to support this pyrolysis comes from the combustion of approximately 9 g/s of biomass). Not all the CO in the fuel gas mixture is consumed in the SOFC. The excess CO is converted to additional H₂ using the biologically based gas conditioning system. This biologically generated hydrogen is used in a PEMFC to electricity.

The biologically based gas conditioning system is an enzymatic CO-water gas shift reaction ($\text{CO} + \text{H}_2\text{O} \rightleftharpoons \text{H}_2 + \text{CO}_2$) employing the phototrophic bacterium *Rubrivivax gelatinosus* CBS. This bacterium can grow on 100% CO to produce near stoichiometric amounts of H₂. Cultures of *Rx. gelatinosus* CBS fed with CO at 20,000 ppm in the culture gas phase (at a flow rate of 500 ml [0.625g] CO/min) reduced the CO to a mere 0.1 ppm at equilibrium. Thus, *Rx. gelatinosus* may be used for both for fuel gas conditioning and for producing H₂.

The flow rate of CO from the gasification system is 0.24 g/s. The rate of CO-oxidation by *Rx. gelatinosus* CBS is 8.3 mmol CO/min/g cdw. Thus *Rx. gelatinosus* CBS can fix CO at a rate of approx. 0.5 mol [= 14.0 g] CO uptake/hr/g cdw. Thus, oxidation of 864 g CO/hr, would require approximaely 62 g *Rx gelatinosus* CBS. An Optical Density of 2.0 yields 1.5 g *Rx. gelatinosus* CBS cdw/liter. Thus, a 41liter culture of *Rx. gelatinosus* CBS at an OD of 2.0 would be sufficient to capture the CO generated by the gasification system. Since hydrogenase reaction relies on the transferring of the reducing equivalents from CO oxidation, via several other intermediates, to its active site, the overall H₂ production rate from CO in general is less than the CO uptake rate reported above. The best rate of H₂ production from CO is 1.5 mmol H₂ produced/min/g cell dry wt., obtained by vigorously shaking a culture of low cell density. Thus, a 41 litre culture (62 g) of *Rx gelatinosus* CBS could produce an additional 5.6 moles H₂/hr. Better reactor design should improve this rate significantly.