

Bio-Derived Liquids to Hydrogen Distributed Reforming Working Group Background Paper

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Key Drivers

Biomass is an abundant renewable resource (over 1 billion dry tons could be available annually) and is a potential feedstock for hydrogen production [1]. Hydrogen can be produced by reforming bio-liquids such as sugars, ethanol, or bio-oils or through gasification or pyrolysis of biomass feedstocks. In the near term, distributed hydrogen production technologies such as bio-liquid reforming may be the most viable renewable hydrogen pathway due to their lower capital investment requirements (including hydrogen transport and delivery infrastructure). The U.S. Department of Energy (DOE) Hydrogen, Fuel Cells and Infrastructure Technologies Program (HFCIT) is cost-sharing research to address technical challenges and lower the cost of producing hydrogen from renewable liquid fuels as part of HFCIT's Hydrogen Production subprogram.

Working Group Purpose

The Bio-Derived Liquids to Hydrogen Distributed Reforming Working Group (BILIWG) will provide a forum for effective communication and collaboration among active participants in DOE-HFCIT cost-shared research directed at distributed bio-liquid reforming. The working group will promote and facilitate the active exchange of knowledge, lessons learned, and other pertinent information of common interest. It will aim to reduce unnecessary duplication of efforts, leverage unique expertise and facilities, and foster partnerships and information sharing that can accelerate the development and deployment of distributed bio-liquid reforming and hydrogen purification technologies.

Working Group Scope

The proposed scope for the BILIWG is addressing technical challenges to distributed reforming of biomass-derived, renewable liquid fuels to hydrogen. This includes the reforming, water-gas shift, and hydrogen recovery and purification steps. A distributed production unit is defined as a 1,500 kg H₂ per day design capacity to be located at the forecourt or point of dispensing to the consumer. The scope of the working group does not include production of the renewable liquid fuels, production of syngas or biomass-derived pyrolysis oils, or production of hydrogen from biomass-derived syngas.

DOE Technical Targets

DOE-HFCIT has established technical targets for distributed production of hydrogen from bio-derived renewable liquids, as shown in Table 1. These targets represent the goals for DOE-sponsored R&D and will be used to measure progress.

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Table 1. Technical Targets: Distributed Production of Hydrogen from Bio-Derived Renewable Liquids^{a,b}

Characteristics	Units	2006 Status ^c	2012 Target ^c	2017 Target ^d
Production Unit Energy Efficiency ^e	%	70.0	72.0	65-75 ^f
Production Unit Capital Cost (Un-installed)	\$	1.4M	1.0M	600K
Total Hydrogen Cost	\$/gge	4.40	3.80	<3.00

- a. These costs are based on modeling the cost of distributed renewable liquids reforming in the H2A "Forecourt Production Modeling Tool" downloadable from http://www.hydrogen.energy.gov/h2a_production.html. Specific assumptions used to achieve the overall hydrogen cost objectives are documented in Record 6003. Record 6003 is posted on (www.hydrogen.energy.gov/program_records.html) for public review.
- b. The H2A Forecourt Production Model was used with following standard economic assumptions: All values are in 2005 dollars, 1500 kg/day design capacity, 1.9% inflation rate, 10% After Tax Return on Investment, 100% Equity Financing, 7 year MACRS depreciation, 20 yr analysis period, 38.9% overall tax rate, 70% capacity factor, and 15% working capital. It is assumed that Design for Manufacture and Assembly (DFMA) would be employed and that on the order of 500 units per year would be produced. The capital cost for the forecourt station compression and storage are consistent with the status and targets in the Delivery Section 3.2. Based on the recommendations made by the 2006 Independent Assessment of the Status of Distributed Natural Gas Reforming (www.eere.energy.gov/hydrogenandfuelcells) start-up time was set to 0.5 years, % variable costs in year 1 was set to 50%, and % fixed cost in year 1 was set to 75%.
- c. The 2006 Status and 2012 Targets are based on the H2A Distributed Ethanol team reforming analyses Current and Advanced cases respectively (www.hydrogen.energy.gov) with respect to the production unit capital and operating efficiency. The cost of ethanol utilized is \$1.07/gal. This is the DOE EERE Biomass Program target for cellulosic based ethanol in 2012. The electricity cost utilized is \$.08/kWh based on the EIA 2005 Annual Energy Outlook High A case projection for 2015 in 2005\$.
- d. The 2017 Target has been set to achieve <\$3.00/gge hydrogen. Aqueous Phase Reforming of sugars is a technology being researched that has the potential to reach this Target and was used as the example H2A Distributed Production case run. The cost of sugar used was \$.07/lb which is consistent with the target cost of cellulosic sugar for ethanol production in 2012 in the DOE EERE Biomass Program. The electricity cost utilized is \$.08/kWh based on the EIA 2005 Annual Energy Outlook High A case projection for 2015 in 2005\$. The capital cost and energy efficiency of the production unit are based on preliminary analyses and projections for what could be achieved with successful development of this technology. (See record 6003, www.hydrogen.energy.gov/program_records.html for more details.) Alternatively, the Target of <\$3.00/gge could be achieved with ethanol reforming if the cost of ethanol could be reduced to <\$.90/gal. This ethanol cost is consistent with the longer term (>2015) DOE EERE Biomass Program cost target for cellulosic ethanol.
- e. Energy Efficiency is defined as the energy in the hydrogen produced (on a LHV basis) divided by the sum of the feedstock energy (LHV) plus all other energy used in the process.
- f. Production Unit Energy Efficiency may vary (as low as 65%) as the capital cost, feedstock costs and other costs associated with aqueous phase reforming is low enough to still achieve the Target of <\$3.00/gge hydrogen cost.

Source: HFCIT FY2007 RD&D Plan

Table 2 provides the breakdown of the cost contributors taken directly from the H2A Forecourt Production Model case (aqueous phase reforming of ethanol) used to generate the targets in Table 1. The values in Table 2 are not targets, but rather demonstrate the potential cost breakdown of the H2A example cases at the specified targets. They highlight the impacts of feedstock and reforming technology choice on hydrogen cost and may help drive R&D needs prioritization.

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Table 2. A Distributed Bio-Derived Renewable Liquids H2A Example - Cost Contributions^a

Characteristics	Units	2006 Status	2012 Target	2017 Target
Production Unit Capital Cost Contribution	\$/gge	0.75	0.45	0.40
Storage, Compression, Dispensing Capital Cost Contribution ^b	\$/gge	0.75	0.55	0.35
Fixed O&M Cost Contribution	\$/gge	0.60	0.50	0.40
Feedstock Cost Contribution	\$/gge	2.10	2.10	1.55
Other Variable O&M Cost Contribution	\$/gge	0.20	0.20	0.30
Total Hydrogen Cost	\$/gge	4.40	3.80	3.00

a. This table provides the breakdown of the cost contributions taken directly from the H2A Forecourt Production Model cases used to generate the targets in Table 1. These are not targets. They provide additional insight into the cost breakdown of these example H2A cases at the specified targets.

b. Storage capacity for 1000 kg of hydrogen at the forecourt is included. It is assumed that the hydrogen refueling fill pressure is 5000 psi for 2003, 2005 and 2010. It is assumed that in 2015, the hydrogen refueling fill pressure is 10,000 psi.

Source: HFCIT FY 2007 RD&D Plan

HFCIT R&D Efforts

HFCIT is currently supporting several distributed bio-liquid reforming research efforts (see Table 3). These technologies target a range of bio-liquid feedstocks, including ethanol, glucose, sorbitol, glycerol, and bio-oil. In addition, lessons learned from HFCIT research on distributed natural gas reforming will be applied to bio-liquid reforming where appropriate.

Bio-Liquid Reforming Feedstocks

Ethanol is the most heavily researched renewable liquid because it is easy to store, handle, and transport due to its low toxicity and volatility [7, 8]. The ethanol production and transportation infrastructure already exists and is undergoing expansion to meet the increasing ethanol demand created by the Energy Policy Act of 2005 and other state legislation [9, 10, and 11].¹ Therefore it appears to be the most viable mid-term approach to bio-liquid reforming. Other renewable liquid options include sorbitol, glucose, glycerol, bio-oil, methanol, propylene glycol, and less refined sugar streams (cellulose, hemicellulose). Table 4 lists the potential bio-liquid feedstocks, reforming technology and feedstock development time frame, feedstock cost, theoretical hydrogen yield, and the individual bio-liquid feedstock advantages and disadvantages.

¹ See addendum for additional information on ethanol production and distribution infrastructure.

Table 3. HFCIT Renewable Bio-Liquids Reforming Research

Project	Partners (PI in Bold)	Technology	Bio-Liquid Feedstocks	Current H2 yield
Hydrogen Generation from Biomass-Derived Carbohydrates via the Aqueous-Phase Reforming Process	Virent Energy Systems, Inc. ; U. of Wisconsin; ADM; Universal Oil Products LLC	Aqueous-phase reforming	Sugars (glucose); sugar alcohols; glycerol	Using 10 wt% feed of: <ul style="list-style-type: none"> Glucose, 3,000 $\mu\text{mol H}_2/\text{g cat/h}$ Sorbitol, 6,000 $\mu\text{mol H}_2/\text{g cat/h}$ Glycerol, 20,000 $\mu\text{mol H}_2/\text{g cat/h}$ Ethylene glycol, 40,000 $\mu\text{mol H}_2/\text{g cat/h}$ Methanol, 40,000 $\mu\text{mol H}_2/\text{g cat/h}$ [2]
Production of Hydrogen by Biomass Reforming	PNNL	Aqueous phase reforming Vapor phase steam reforming	Sorbitol Ethanol	<ul style="list-style-type: none"> Highest productivity of 141 L H₂/L catalyst/hr (10% sorbitol feed, 265°C, 835 psi, 3%Pt/Al₂O₃) [3]
High-Pressure Distributed Ethanol Reforming	ANL	High pressure steam reforming.	Ethanol	At S/C = 6, P = 2000 psia [4] <ul style="list-style-type: none"> T=800°C, H₂ yield ~54% T=900°C, H₂ yield ~72% T=1000°C, H₂ yield ~80%
Investigation of Reaction Networks and Active Sites in Bio-Ethanol Steam Reforming over Co-Based Catalysts	Ohio State University	Low temperature (350-550°C) steam reforming of ethanol using non-precious metal (cobalt) catalyst system.	Ethanol	<ul style="list-style-type: none"> H₂ yields >70% at GHSV^a approaching 100,000 hr⁻¹ (EtOH:H₂O = 1:10 molar) [5]
Distributed Bio-Oil Reforming	NREL	Whole bio-oil is revolatized, 10% methanol added to stabilize the oil, and reformed in a fluid bed reactor	Bio-oil (36.5% carbon, 8.4% hydrogen, 55.0% oxygen)	<ul style="list-style-type: none"> Commercial catalyst: 12.9 g H₂/100 g bio-oil (without water-gas shift)^b NREL catalysts: lower H₂ yields than commercial catalyst, but with much lower catalyst attrition [6]

^a Gas hourly space velocity.

^b This experiment did not include the water-gas shift. The yield is expected to increase by 10% if the water-gas shift step was added.

Table 4. Potential Bio-Liquid Reforming Pathways				
Bio-Liquid	Time Frame ^{a, b}	Bio-Liquid Cost (Plant-Gate)	Theoretical H2 Yield	Feedstock Advantages/ Disadvantages
Ethanol	Mid-term	\$1.07/gallon ^c	0.78 kg H2/ gallon ethanol 0.26 kg H2/kg ethanol	<ul style="list-style-type: none"> ▪ Low toxicity ▪ Low sulfur content ▪ Use of dilute ethanol would reduce reforming feedstock costs ▪ Ethanol production/ delivery infrastructure is already established
Glucose	Mid-term	\$0.07/lb ^d	0.13 kg H2/kg glucose	<ul style="list-style-type: none"> ▪ Low volatility ▪ Non-toxic, non-flammable
Glycerol	Mid-term	\$0.15/lb (80% glycerol, ~20% water from biodiesel production) ^e [12]	0.15 kg H2/kg pure glycerol	<ul style="list-style-type: none"> ▪ Utilizes low-value glycerol by-product from biodiesel production ▪ Low volatility ▪ Non-toxic, non-flammable
Crude Glycerol (CG)	Mid-term	<\$0.15/lb ^f	23.6 g H2/ 100 g CG [13] 0.24 kg H2/ kg CG	<ul style="list-style-type: none"> ▪ Low volatility ▪ High reactivity; potential of forming carbonaceous deposits or converting to aromatics that are more difficult to reform to H2 [6]
Bio-oil	Mid-term	\$0.03-0.04/ lb bio-oil ^g	13.8 g H2/100 g bio-oil [6] 0.06 kg H2/ lb bio-oil	
Sorbitol	Long-term	\$0.10/lb ^h	0.13 kg H2/kg sorbitol	<ul style="list-style-type: none"> ▪ Low volatility ▪ Non-toxic, non-flammable
Ethylene Glycol (EG) and Propylene Glycol (PG)	Long-term	EG: \$0.44-0.46/lb [14] ⁱ PG: \$0.71-1.02/lb [15] ⁱ	0.15 kg H2/kg EG 0.22 kg H2/kg PG	<ul style="list-style-type: none"> ▪ Low volatility ▪ Non-toxic (PG), non-flammable (both)
Cellulose/ Hemicellulose	Long-term	\$0.07/lb ^j	~0.13 kg H2/kg cellulose- hemicellulose	<ul style="list-style-type: none"> ▪ Low volatility ▪ Non-toxic, non-flammable
Methanol	Long-term	\$0.78-0.91/gallon ⁱ [16]	0.64 kg H2/gallon methanol 0.22 kg H2/kg methanol	<ul style="list-style-type: none"> ▪ More easily reformed to hydrogen than ethanol ▪ High toxicity ▪ Higher corrosivity, volatility than ethanol

^a Near-term (2012), mid-term (2012-2017), long-term (2017+)

^b This is based on the market readiness of both the reforming technology and the bio-liquid production and distribution infrastructure.

^c This is the DOE EERE Biomass Program target for cellulosic ethanol in 2012.

^d This is glucose price from the 2004 H2A Central Sorbitol Production analysis which assumes an nth plant biorefinery with glucose as one of the product streams.

^e Methanol, fatty acids, and most of the water have been removed [12].

^f Assumes the cost is of crude glycerol is lower than semi-purified glycerol. Crude glycerol is defined as 55% glycerol and 45% methyl esters of fatty acids [13].

^g This represents the Office of the Biomass Program 2010 and 2020 pyrolysis oil production cost goals of \$5.10/MM Btu and \$4.30/MM Btu, respectively [17]. Bio-oil energy content is assumed to be 7,500 Btu/lb [18].

^h This is from the 2004 H2A Central Sorbitol Production analysis (2000\$) using \$0.07/lb glucose.

ⁱ This is the cost of the fossil derived product. The biobased product will have to be cost-competitive.

^j Consistent with the target cost of cellulosic sugar for ethanol production in 2012 in the DOE EERE Biomass Program.

As Table 4 shows, ethanol has the highest theoretical hydrogen yield per pound of feedstock. Based on current hydrogen yields from ethanol steam reforming and the potential biomass resources available domestically (1 billion dry tons annually), ethanol reforming could supply nearly 64 billion kg H₂ annually on an energy content basis.² This represents approximately 45% of our 2005 gasoline consumption.³

Key Reforming Technical Challenges

The ability of bio-liquid reforming to meet the demand for clean, cost-competitive renewable fuel will require advances in catalyst systems, purification technologies, and integration of these technologies into a single process. A multitude of catalyst systems have been investigated for the steam reforming of ethanol, bio-oil, sugar alcohols, and other bio-liquids [8, 19, 20]. A common problem with the catalysts reviewed is deactivation due to coking which occurs when side reaction products (e.g., acetaldehyde, ethylene) deposit on the catalyst [7, 8, 19]. To a certain degree, process parameters such as the steam-to-carbon (water-to-ethanol) ratio can be modified—in this case increased—to limit carbon deposits, but at the cost of greater process energy requirements [7, 19, 21, 22]. The water-to-ethanol feed ratio and operating temperature also influence the selectivity to hydrogen. Water-to-ethanol molar ratios of three or greater, and temperatures above 500°C have been shown to favor the production of hydrogen over methane and other reaction intermediates [19, 21, 22]. However, higher water-to-ethanol ratios will require increased energy inputs.

Low-temperature (<500°C) reforming technologies are also under investigation. The advantages of low-temperature technologies are reduced energy intensity, compatibility with membrane separation, favorable conditions for water-gas shift reaction, and minimization of undesirable decomposition reactions typically encountered when carbohydrates are heated to high temperatures [3, 23]. Aqueous-phase reforming is a promising technology that has been applied to glucose, ethylene glycol, sorbitol, glycerol, methanol [2, 20, 24]. Studies have shown that the following factors promote selectivity to hydrogen rather than alkanes [20]:

- catalysts made of platinum, palladium, and nickel-tin (nickel catalysts favor alkane production)
- more basic catalyst support materials (e.g., alumina)
- neutral and basic aqueous solutions
- feedstock type (in descending order of hydrogen selectivity) – polyols (selectivity decreases with increasing carbon number), glucose (selectivity decreases as weight % increases from 1 to 10).

Catalyst coking and deactivation are not significant problems as they are in steam reforming, but that may be the result of differences in feedstock reaction pathways. While hydrogen yields are highest from the aqueous phase reforming of sorbitol, glycerol, and ethylene glycol, glucose reforming which has low hydrogen yields may be more practical. Improvements in catalyst performance, reactor design, and reaction conditions may help increase hydrogen selectivity [2]. Low-temperature gas phase reforming of ethanol is also being investigated, but there is a tradeoff between catalyst activity and resistance to deactivation (due to coking) and research is ongoing [3].

² The calculation assumes the following: 1) an average hydrogen yield of 5.4 mole H₂ per mole ethanol [8]; 2) an average ethanol yield of 90 gallons of ethanol per dry ton biomass [25]; 3) biomass feedstock availability of 1 billion dry tons annually [1]; and 4) a gallon gasoline equivalent of 1kg H₂ (ignores increased efficiency of fuel cells compared to gasoline internal combustion engines).

³ 2005 gasoline consumption was approximately 8,933,000 barrels per day [26].

Hydrogen Recovery and Purification Technology Status

Hydrogen purity is critical as the carbon monoxide (CO) must be less than 0.2 ppm to avoid fuel cell degradation [27]. Pressure swing adsorption (PSA), membrane separation, cryogenic distillation, or methanation can be used to lower the CO content of the hydrogen product [8, 28, 29]. The characteristics of these technologies are as follows [28]:

- PSA: commercially-available, able to produce high-purity hydrogen (up to 99.99%); DOE-funded research has achieved the DOE 2005 target for hydrogen purification/separation efficiency of 82% [30]
- Membrane Separation: commercially-available, low energy consumption, relatively low investment cost
- Cryogenic Distillation: commercially-available, high energy consumption, high hydrogen at moderate purities (95% H₂ or less) is possible, but very high purity is not practical
- Methanation: consumes some of the hydrogen product to convert CO to methane.

Of the four technologies, membrane technologies may have the greatest potential for forecourt hydrogen purification due to their low energy consumption and capital investment. Catalysts for reforming and water-gas shift reactions could be incorporated into the membrane so that the reversible reforming/water-gas shift reactions shift to the right, favoring hydrogen production. This would enable the reforming and separation to be performed in one unit, reducing capital costs. Although the use of both inorganic (e.g., metal, ceramics) and organic (polymers) membranes has been investigated, much of current research focuses on metal and ceramic membrane technologies due to their ability to tolerate harsher conditions than organic membranes [28].

Further Considerations:

- 1) What is the technology transfer potential for distributed natural gas reforming technologies?
- 2) What are the pros and cons of fuel flexible reformers?
- 3) What are the impacts of bio-derived liquid feedstock impurities on pressure swing adsorption and hydrogen membranes?
- 4) What is the optimal Water-Gas-Shift technology for bio-derived liquid reforming?
- 5) What are the gaps in catalyst development for bio-derived liquid reforming?
- 6) Are lower system efficiencies for aqueous phase reformers (compared to distributed natural gas reformers) acceptable given lower capital costs?

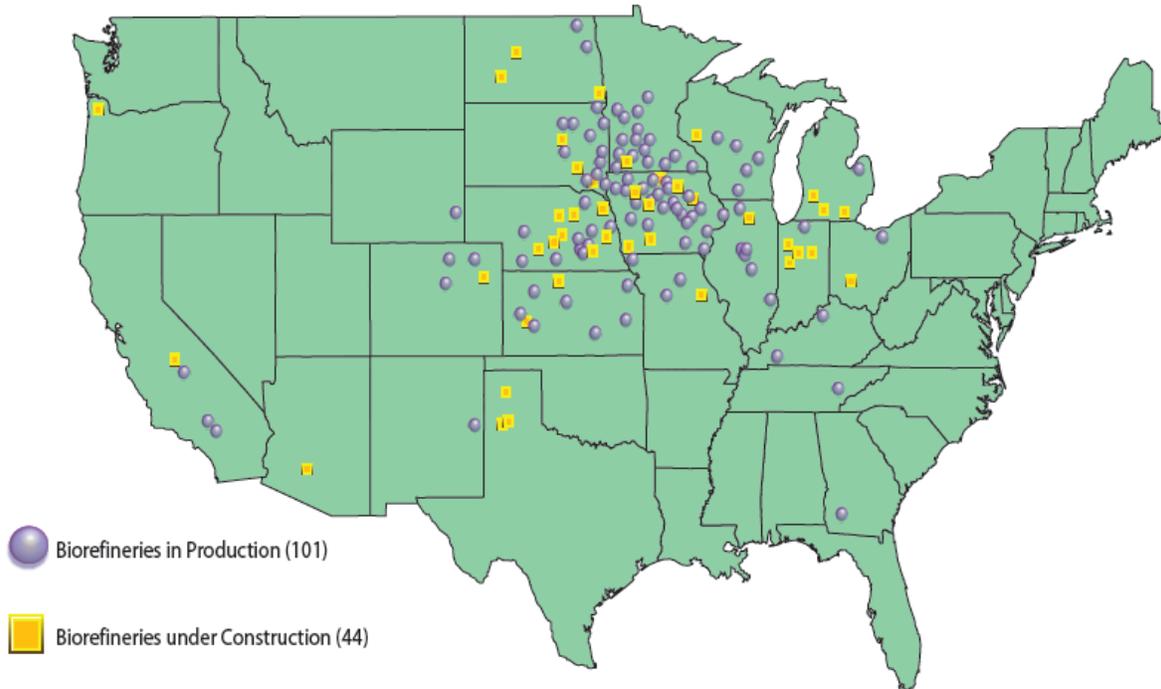
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ADDENDUM

U.S. Ethanol Biorefinery Locations



Source: Renewable Fuels Association, 2006, <http://www.ethanolrfa.org>.

2005 Ethanol Production Capacity: 6.3 billion gallons per year (excluding capacity under construction/expansion of 2.0 billion gallons per year) [11].

Ethanol transportation/distribution occurs by: barge (30-35%), rail (30-35%), and truck (30-35%) [31].