Supplementary information

The environmental biorefinery: state of the art on the production of hydrogen and value-added biomolecules in mixed-culture fermentation

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Table S1: Technology readiness level (TRL) scale according to the European Commission

<table>
<thead>
<tr>
<th>TRL</th>
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<tr>
<td>1</td>
<td>Basic principles observed</td>
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<tr>
<td>2</td>
<td>Technology concept formulated</td>
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<td>3</td>
<td>Experimental proof of concept</td>
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<td>4</td>
<td>Technology validated in lab</td>
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<tr>
<td>5</td>
<td>Technology validated in relevant environment (industrially relevant environment in the case of key enabling technologies)</td>
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<tr>
<td>6</td>
<td>Technology demonstrated in relevant environment (industrially relevant environment in the case of key enabling technologies)</td>
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<td>7</td>
<td>System prototype demonstration in operational environment</td>
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<td>8</td>
<td>System complete and qualified</td>
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<td>9</td>
<td>Actual system proven in operational environment (competitive manufacturing in the case of key enabling technologies)</td>
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Table S2: Patent database

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<th>Priority date</th>
<th>Title</th>
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<tr>
<td>2015</td>
<td>Food waste fermentation device using soil microbes and livestock manure composting device, biofield generator suitable for same, microbial cell suitable for same and microbial condenser suitable for same</td>
<td>WO2016129846 A1;</td>
<td>Application (WO)</td>
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<td>2015</td>
<td>Processing of biomass materials</td>
<td>WO2016160955 A1;</td>
<td>Application (WO)</td>
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<td>2014</td>
<td>Conversion of biomass, organic waste and carbon dioxide into synthetic hydrocarbons</td>
<td>US2016186072 A1; WO2016101076 A1;</td>
<td>Application (US; WO)</td>
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<td>2014</td>
<td>Maximizing production of hydrogen from waste materials by active removal of hydrogen</td>
<td>WO2016040074 A1;</td>
<td>Application (WO)</td>
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<td>2014</td>
<td>Device for high-efficiency biological hydrogen and methane production</td>
<td>TW201602017 A;</td>
<td>Application (TW)</td>
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<td>2014</td>
<td>Method for producing hydrogen by means of dark fermentation from biomass from the wine-producing industry, without using a microbial consortium</td>
<td>WO2016030623 A1;</td>
<td>Application (WO)</td>
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<td>2014</td>
<td>Processes for controlling the concentration of co-produced oxygenated organics in anaerobic fermentation broths for the bioconversion of syngas to product oxygenated organic compound</td>
<td>US2016010123 A1; WO2016007659 A1;</td>
<td>Application (US; WO)</td>
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<td>2014</td>
<td>Microorganism co-culture system and uses of the same</td>
<td>US2016068919 A1; CN105400860 A; TW201610165 A;</td>
<td>Application (US; CN; TW)</td>
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<td>2014</td>
<td>Wastewater treatment method</td>
<td>TW201545995 A; Application (TW)</td>
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<td>2014</td>
<td>The production of hydrogen and other gaseous or liquid products in an accelerated bioprocess</td>
<td>CA2944425 A1; WO2015158950 A1; Application (CA; WO)</td>
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<td>2014</td>
<td>Process for producing biohydrogen and biomethane</td>
<td>MDA436 C1; Active (MD 2016)</td>
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<td>2014</td>
<td>Process for phototrophic production of phas and h2 gas from wide range of organic waste</td>
<td>WO2015123522 A1; Application (WO)</td>
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<td>2014</td>
<td>A method for improving the methane production rate through acidification pretreatment of hydrogen production of kitchen waste and sludge</td>
<td>CN104561222 A; WO2015143906 A1; Application (CN; WO)</td>
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<td>2013</td>
<td>Facilities for resource-recycling biotechnical hydrogen production from biomass</td>
<td>JP2015089945 A; Application (JP)</td>
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<td>2013</td>
<td>Biohydrogen production method and reactor</td>
<td>US2015111273 A1; CA2926577 A1; WO2015058295 A1; BR112016008913 A1; AU2014339713 A1; SG11201663032X A1; KR20160608965 A; CN10572986 A; IL245228 A; EP3060672 A1; IN20167051590 A; VN48590 A; CU20160053 A7; Application (US; CA; WO; BR; AU; SG; KR; CN; IL; IN; CU) Withdrawn (EP 2017)</td>
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<td>2013</td>
<td>Circulatoy bio hydrogen production facility using biomass</td>
<td>JP2015025172 A; Application (WO)</td>
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<td>2013</td>
<td>Bioelectrochemical system having polyvalent ion removing function</td>
<td>WO20141956825 A1; EP3006407 A1; CN105555751 A; US2016137535 A1; JP2016532441 A; Application (WO; EP; CN; US)</td>
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<td>2013</td>
<td>Hydrogen production from dark fermentation of high-protein material under anaerobic condition</td>
<td>KR20140112292 A; KR101472379 B1; Active (KR 2014)</td>
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<td>2013</td>
<td>Method and plant for continuously producing hydrogen (h2) and methane (ch4) from zootechnical effluents</td>
<td>WO2014147558 A1; EP2976430 A1; Application (WO; EP)</td>
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<td>2013</td>
<td>Digestor assemblies to provide renewable resources</td>
<td>US2014273196 A1; WO2014160252 A1; US912724 B2; Application (WO) Active (US 2015)</td>
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<td>2013</td>
<td>Method for production of n-propanol and other c3-carbon containing products from syngas by symbiotic arrangement of c1-fixing and c3-producing anaerobic microorganism cultures</td>
<td>US2014273123 A1; WO2014140336 A1; CN105729987 A; Application (US; WO; CN)</td>
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<td>2012</td>
<td>Production of hydrogen using an anaerobic biological process</td>
<td>US2014157777 A1; US9506084 B2; Active (US 2016)</td>
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<td>2012</td>
<td>Microbial consortium for the production of hydrogen</td>
<td>WO2014033345 A1; ES2456776 B1; Application (WO)</td>
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<td>2012</td>
<td>Microbial electrolysis cells and methods for the production of</td>
<td>US2013256149 A1; US9216919 B2; Active (US 2015)</td>
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2012 Process for anaerobic production of biohydrogen from organic waste MD4217 C1; Active (MD 2013)

2012 Method for producing lactic acid using continuous anaerobic reactors KR101271005 B1; Active (KR 2013)


2012 Method and system for electro-assisted hydrogen production from organic material CA2860463 A1; US2013217089 A1; WO2013120206 A1; AU2013220906 B2; BR112014020962 A1; CN104245944 A; EP2814971 A4; IN7590DEN2014 A; US9458474 B2; US2016333379 A1; Application (CA; WO; BR; CN; EP; IN) Active (US 2016; AU 2016)


2011 Development of a combined biohydrogen and methane production unit using two-stage anaerobic co-digestion process IN4505CHE2011 A; Application (IN) Active (KR 2012)

2011 Method and apparatus for anaerobically digesting organic material US2013137153 A1; Application (US)


2011 Pre-thermal treatment of microalgae and high temperature and high efficiency hydrogen/methane fermentation process using waste heat of power-plant effluent gas KR101181834 B1; Active (KR 2012)


2011 Apparatus for producing biohydrogen and biomethane from organic substance LV14431 B; Active (2012)

2011 Method and apparatus for manufacturing a hydroxycarboxylic acid product and use of bleaching filtrate FIE20154596 A; WO2012160255 A1; UY34085 A; Application (FI; WO; UY)

2010 PROCESS FOR PRODUCTION OF HYDROGEN AND VOLATILE GREASY ACIDS BRP10005215 A8; -

2010 Methods and systems for the production of alcohols and/or acids CA2789333 C; CA2836686 C; WO2012054806 A3; TW201231669 A; AU2011316899 B2; US2011312576 A1; CN103298753 B; JP2013521802 A; BR112013003609 A2; KR20150141194 A; Application (CA; WO; BR)


2010 Method for producing energy-rich gases from lignocellulosic material streams WO2012003556 A1; BR11201201605 A2; Application (WO, BR)

2010 Continuous Process and device of production of ethanol from [organico] garbage and other residues of biomass BRP10000194 A2; Application (BR)


2009 A bioelectrochemical cell system WO2010111829 A1; AU2010278674 A1; Application (WO; AU)


2009 Bio-electrochemical device and method for upgrading a fluid NL2003812 C; WO2011062485 A3; EP2502301 A2; Application (WO)

2009 A method for dihydrogen gas production TW201107480 A; TWI406949 B; Application (TW 2013)

2009 Methods for selectively producing hydrogen and methane US2011033908 A1; WO20120217902 A1; Application (WO; AR)
from biomass feedstocks using an anaerobic biological system AR077702 A1; US8246828 B2; Withdrawn (US 2016)


2009 Diphasic algal culture system WO2010150190 A3; AU2010264093 B2; IN1716MUMNP2012 A; CN102575209 B; ZA201200430 B; Application (WO; IN) Active (CN 2016; ZA 2014) Withdrawn (AU 2018)


2009 Method for energetic, waste-free recycling of vegetable raw materials, which are converted into energy source e.g. ethanol and methane and into a fertilizer, comprises hydrolyzing vegetable raw materials, and fermenting DE102009024423 A1; Withdrawn (DE 2012)


2009 Integrated system for hydrogen and methane production from industrial organic wastes and biomass CA2751046 A1; VO201005893 A1; AU2010207087 B2; EP3291706 A4; CN102300977 B; US201209643 A1; IN1716MUMNP2011 A; BRP11007417 A2; US8900840 B2; US2014370587 A1; US2016066794 A1; US9303242 B2; Application (WO; AU; TR; CN; VN; JP; BR; TH; RU) Active (CA 2017; AU 2015; CN 2015; US 2014)

2009 Compositions and methods for conversion of lignocellulosic material to fermentable sugars and products produced therefrom CA2755449 C; WO2010107944 A1; US2012026673 A1; EP2408924 A1; TR201109113 T1; CN102449156 A; VN29679 A; MX2011009745 A; JP2012520682 A; BRP11012700 A2; TH120309 A; RU2011139512 A; US8658407 B2; Application (WO; AU; TR; CN; VN; JP; BR; TH; RU) Active (CA 2015; US 2014) Withdrawn (EP 2013)


2008 Production of methanol or methanol derivatives WO2010068979 A1; Application (WO)

2008 Alcohol production process US2009275787 A1; US8119844 B2; Application (WO)


2008 Method and device for microbial production of a certain product and methane TW200936762 A; TW1354704 B; Application (WO)

2008 Production of hydrogen and/or ethanol by the anaer TW200936762 A; TW1354704 B; Active (TW 2011)


2008 Systems and methods for anaerobic digestion and collection of products GB0915377 D0; JP2464585 B2; US200930805 B2; CA2741199 C; WO201003308 A3; AP201005689 D0; EP2361229 A4; CN102227383 B; HK1143609 A1; US2014315475 A1; AP3504 A1; Application (WO; AP; EP; HK) Active (GB 2012; AU 2014; CA 2016; CN 2014)

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<tr>
<td>2007</td>
<td>Microbial hydrogen-producing process and system thereof</td>
<td>US2009035812 A1; TW200907051 A; US8003344 B2;</td>
<td>Active (US 2011); Rejected (TW 2011)</td>
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<td>2007</td>
<td>Alcohol production process</td>
<td>WO2008115080 A1; NZ553984 A; US2010105115 A1; US8293509 B2;</td>
<td>Application (WO)</td>
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<td>2007</td>
<td>Apparatus and method for biohydrogen production</td>
<td>GB0705553 D0; US2008233624 A1;</td>
<td>Withdrawn (GB 2011; US 2011)</td>
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<td>2006</td>
<td>Device for the combined production of hydrogen and methane by fermentation of biological starting materials, comprises fermentation container, and units for discharge of liquid conversion products and for restraint of micro organisms</td>
<td>DE102006035213 B4;</td>
<td>Active (DE 2012)</td>
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<tr>
<td>2006</td>
<td>A novel treatment method for wastewater with a high concentration of organics and its equipment</td>
<td>TW2007042737 A;</td>
<td>Rejected (TW 2010)</td>
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<td>2006</td>
<td>Process for enhancing anaerobic biohydrogen production</td>
<td>US7232669 B1; TW200734464 A; TW1307364 B;</td>
<td>Active (US 2007; TW 2009)</td>
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<td>2005</td>
<td>Method for obtaining hydrogen and methane from bioresidues, includes two phases, where in former phase aqueous suspension of bioresidues is introduced in reactor, and crushed to obtain particle size less than five millimeter</td>
<td>ES2292312 B1;</td>
<td>Active (ES 2009)</td>
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<td>2005</td>
<td>Process for enhancing anaerobic biohydrogen production</td>
<td>TW1271380 B; TW200724483 A;</td>
<td>Active (TW 2007)</td>
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<td>2005</td>
<td>Hydrogen generating system</td>
<td>JP2007159457 A;</td>
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<td>2005</td>
<td>Two-stage fermentation process and assembly to generate hydrogen and methane</td>
<td>DE102005012367 A1;</td>
<td>Rejected (DE 2009)</td>
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<td>2005</td>
<td>Treatment method of organic waste accompanied by hydrogen production by microorganisms</td>
<td>JP2006223962 A;</td>
<td>Rejected (JP 2010)</td>
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<td>2004</td>
<td>Process for microbial production of hydrogen and methane fuels</td>
<td>US7575907 B1;</td>
<td>Active (US 2009)</td>
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<td>2004</td>
<td>Nutrient salt formula for hydrogen production of anaerobic organism</td>
<td>TW200606127 A;</td>
<td>Rejected (TW 2008)</td>
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<td>2004</td>
<td>Hydrogen fermentation method</td>
<td>JP2005211782 A;</td>
<td>Rejected (JP 2009)</td>
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<td>2004</td>
<td>Hydrogen production apparatus</td>
<td>JP2005200283 A;</td>
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<td>2003</td>
<td>Microbial production of hydrogen under anaerobic condition</td>
<td>TW200417533 A; TW1256946 B;</td>
<td>Active (TW 2006)</td>
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<td>2002</td>
<td>Improved biological production of hydrogen and co-production of methane</td>
<td>TW200404046 A; SG111965 A1; TW138904 B;</td>
<td>Active (TW 2009)</td>
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<td>2001</td>
<td>Method for producing hydrogen by microbial group and the same hydrogen-fermenting microbial group</td>
<td>JP20022272491 A; JP3885868 B2;</td>
<td>Active (JP 2007)</td>
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<td>1999</td>
<td>Production of hydrogen and/or methane of plant body</td>
<td>JP2000232891 A; JP3739584 B2;</td>
<td>Withdrawn (JP 2015)</td>
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The patent status column was assessed in March 2018. Years indicated near country abbreviations in this column correspond to the date of the last status update.
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<td>2017</td>
<td>Effects of pH and substrate concentrations on dark fermentative biohydrogen production from xylose by extreme thermophilic mixed culture</td>
<td>10.1007/s11274-016-2178-1</td>
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<td>2017</td>
<td>Hydrogen production from starch by co-culture of Clostridium acetobutylicum and Rhodobacter sphaeroides in one step hybrid dark- and photofermentation in repeated fed-batch reactor</td>
<td>10.1016/j.biortech.2016.10.060</td>
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<td>2017</td>
<td>Long-term operation of microbial electrolysis cell reducing CO2 to multi-carbon chemicals with a mixed culture avoiding methanogenesis</td>
<td>10.1016/j.biolechem.2016.09.001</td>
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<td>2016</td>
<td>High-purity propionate production from glycerol in mixed culture fermentation</td>
<td>10.1016/j.biortech.2016.08.026</td>
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<td>2016</td>
<td>Regulation of acidogenic metabolism towards enhanced short chain fatty acid biosynthesis from waste: Metagenomic profiling</td>
<td>10.1039/c5ra24254a</td>
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<tr>
<td>2016</td>
<td>Cell wash-out enrichment increases the stability and performance of biohydrogen producing packed-bed reactors and the community transition along the operation time</td>
<td>10.1016/j.renene.2016.05.082</td>
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<td>2016</td>
<td>The type of carbohydrates specifically selects microbial community structures and fermentation patterns</td>
<td>10.1016/j.biortech.2016.09.084</td>
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<td>2016</td>
<td>Dark fermentative hydrogen and ethanol production from biodiesel waste glycerol using a co-culture of Escherichia coli and Enterobacter sp.</td>
<td>10.1016/j.fuel.2016.08.043</td>
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<td>2016</td>
<td>Mesophilic and Thermophilic Biohydrogen Production from Xylose at Various Initial pH and Substrate Concentrations with Microflora Community Analysis</td>
<td>10.1021/acse.energ.yfuels.5b02143</td>
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<td>2016</td>
<td>High-rate hydrogen fermentation of cellulose-lignin fraction in de-oiled jatropha waste using hybrid immobilized cell system</td>
<td>10.1016/j.fuel.2016.05.088</td>
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<td>2016</td>
<td>Short-chain fatty acids production and microbial community in sludge alkaline fermentation: Long-term effect of temperature</td>
<td>10.1016/j.biortech.2016.03.138</td>
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<td>2016</td>
<td>Performance and microbial community analysis in alkaline two-stage enhanced anaerobic sludge digestion system</td>
<td>10.1016/j.bej.2015.10.004</td>
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<td>2016</td>
<td>Microbial responses to various process disturbances in a continuous hydrogen reactor fed with galactose</td>
<td>10.1016/j.jbiosc.2016.08.006</td>
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<td>2016</td>
<td>Evaluation of different pretreatments on organic matter solubilization and hydrogen fermentation of mixed microalgae consortia</td>
<td>10.1016/j.jhydene.2016.05.195</td>
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<td>2016</td>
<td>Changes in microbial community during hydrogen and methane production in two-stage thermophilic anaerobic co-digestion process from biowaste</td>
<td>10.1016/j.wasman.2016.01.016</td>
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<td>2016</td>
<td>Pretreatment conditions of rice straw for simultaneous hydrogen and ethanol fermentation by mixed culture</td>
<td>10.1016/j.jhydene.2015.10.147</td>
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<td>2016</td>
<td>Process enhancement of hydrogen and methane production from palm oil mill effluent using two-stage thermophilic and mesophilic fermentation</td>
<td>10.1016/j.jhydene.2016.05.037</td>
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<td>2016</td>
<td>Co-generation of biogas and biomethane through two-stage batch co-fermentation of macro- and micro-algal biomass</td>
<td>10.1016/j.biortech.2016.06.092</td>
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<td>2016</td>
<td>Optimum alcohol concentration for chain elongation in mixed-culture fermentation of cellulose substrate</td>
<td>10.1002/bit.26024</td>
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<td>2016</td>
<td>Two-stage thermophilic fermentation and mesophilic methanogen process for biohythane production from palm oil mill effluent with methanogenic effluent recirculation for pH control</td>
<td>10.1016/j.jhydene.2016.07.095</td>
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<td>2016</td>
<td>Potential and optimization of two-phase anaerobic digestion of oil refinery waste activated sludge and microbial community study</td>
<td>10.1038/srep38245</td>
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<td>2016</td>
<td>Fermentative biohydrogen and biomethane co-production from mixture of food waste and sewage sludge: Effects of physicochemical properties and pH ratios on fermentation performance</td>
<td>10.1016/j.apenergy.2016.10.003</td>
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<td>2016</td>
<td>Boosting dark fermentation with co-cultures of extreme thermophiles for biohythane production from garden waste</td>
<td>10.1016/j.biortech.2016.07.096</td>
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<td>2016</td>
<td>High-rate hydrogen production from galactose in an upflow anaerobic sludge blanket reactor (UASBr)</td>
<td>10.1039/cr0a09299e</td>
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<td>2016</td>
<td>Critical analysis of hydrogen production from mixed culture fermentation under thermophilic condition (60°C)</td>
<td>10.1007/s00253-016-7482-z</td>
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<td>2016</td>
<td>Production of Bio-Based Hydrogen Enriched Methane from Waste Glycerol in a Two Stage Continuous System</td>
<td>10.1007/s12649-016-9538-9</td>
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<td>2016</td>
<td>Consistent 1,3-propanediol production from glycerol in mixed culture fermentation over a wide range of pH</td>
<td>10.1186/s13068-016-0447-8</td>
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<td>2016</td>
<td>Single-stage and two-stage anaerobic digestion of extruded lignocellulosic biomass</td>
<td>10.1016/j.apenergy.2016.10.039</td>
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<td>2016</td>
<td>Comparison of various carbohydrates for hydrogen production in microbial electrolysis cells</td>
<td>10.1080/13102818.2015.1081078</td>
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<td>2016</td>
<td>Increased performance of hydrogen production in microbial electrolysis cells under alkaline conditions</td>
<td>10.1016/j.biolechem.2016.01.003</td>
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<td>2016</td>
<td>Fermentative hydrogen production in an up-flow anaerobic biofilm reactor inoculated with a co-culture of Clostridium acetobutylicum and Desulfurobacter vulgaris</td>
<td>10.1016/j.biortech.2016.09.072</td>
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<td>2016</td>
<td>Microbial dynamics of the extreme-thermophilic (70 °C) mixed culture for hydrogen production in a chemostat</td>
<td>10.1016/j.jhydene.2016.04.085</td>
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<td>2016</td>
<td>Towards biohythane production from biomass: Influence of operational stage on anaerobic fermentation and microbial community</td>
<td>10.1016/j.jhydene.2015.06.045</td>
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<td>2016</td>
<td>Fermentative production of hydrogen in presence of modified mesoporous silica SBA-15</td>
<td>10.1016/j.ijhydene.2016.05.112</td>
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