

# Chemical Biorefinery

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“A biorefinery is an integrated system with efficient and flexible conversion of biomass feedstocks, through a combination of physical, chemical, biochemical and thermochemical processes, into multiple products.”

*Biorefineries and Chemical Processes: Design, Integration and Sustainability Analysis*, First Edition.

Jhuma Sadhukhan, Kok Siew Ng and Elias Martinez Hernandez.

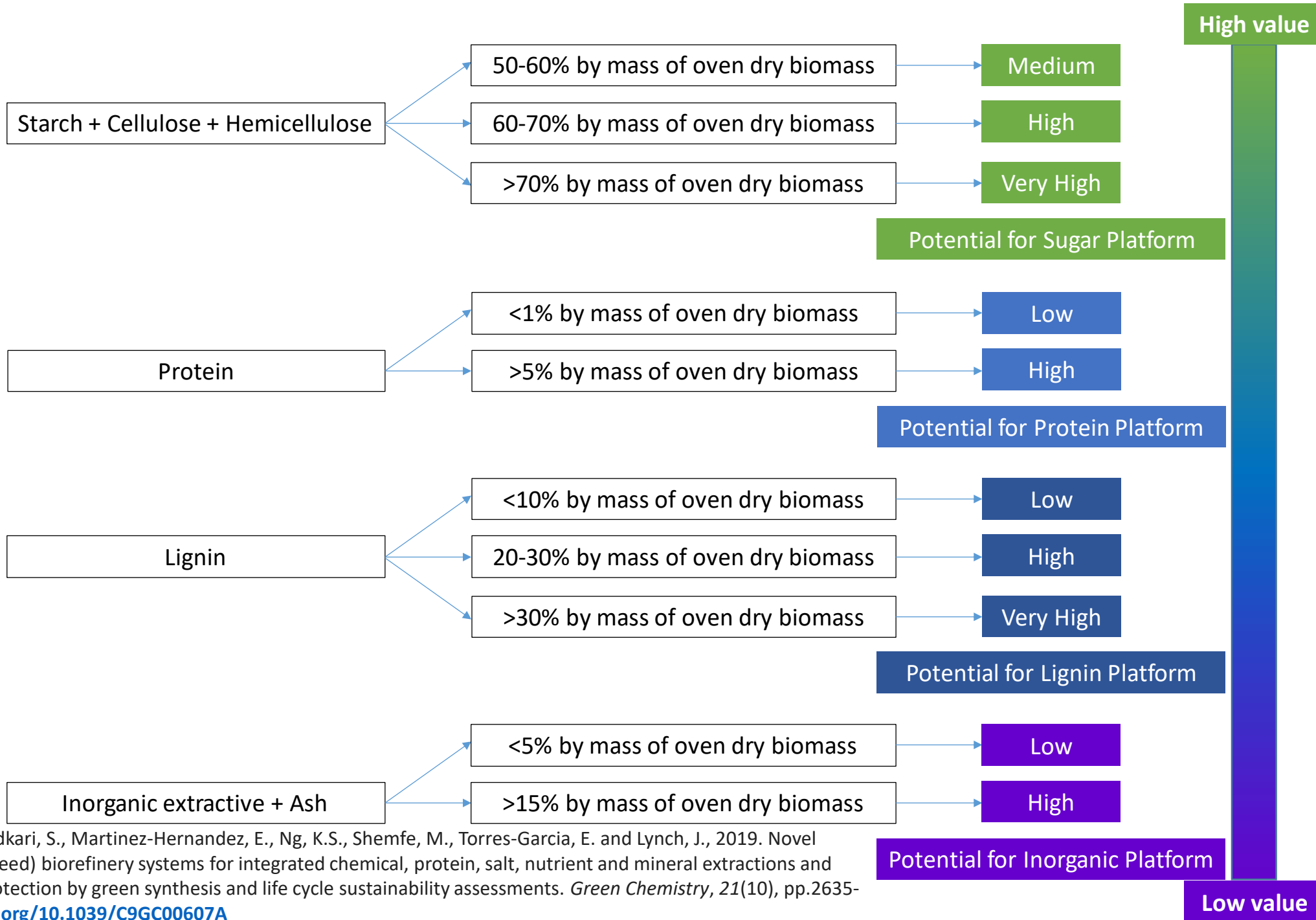
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Companion Website: <http://www.wiley.com/go/sadhukhan/biorefineries>

% mass of dry biomass	Brown	Green	Red	Hardwood	Softwood
	(Sargassum)	(Sea lettuce)	(Euchemia)		
Starch	0.1	0.7	1	0.5	0.1
Cellulose	20.3	8	6	43.9	37.9
Hemicellulose	42.8	42.1	66	28.4	22.7
Lignin	7.3	3.3	1.8	24	33.1
Protein	9.6	12	7.5	0.6	0.5
Extractives	1.9	4.1	1	1.9	3.4
Ash	17.1	25.7	15	0.6	0.3
Total	99.1	95.9	98.3	99.9	98
Calorific value (MJ kg <sup>-1</sup> )	14.02	15.88	12.5	20.62	19.35
Potential for extraction of platform:					
Sugar	High	Medium	Very high	Very high	High
Protein	High	High	High	Low	Low
Lignin	Low	Low	Low	High	Very high
Extractives and ash	High	High	High	Low	Low

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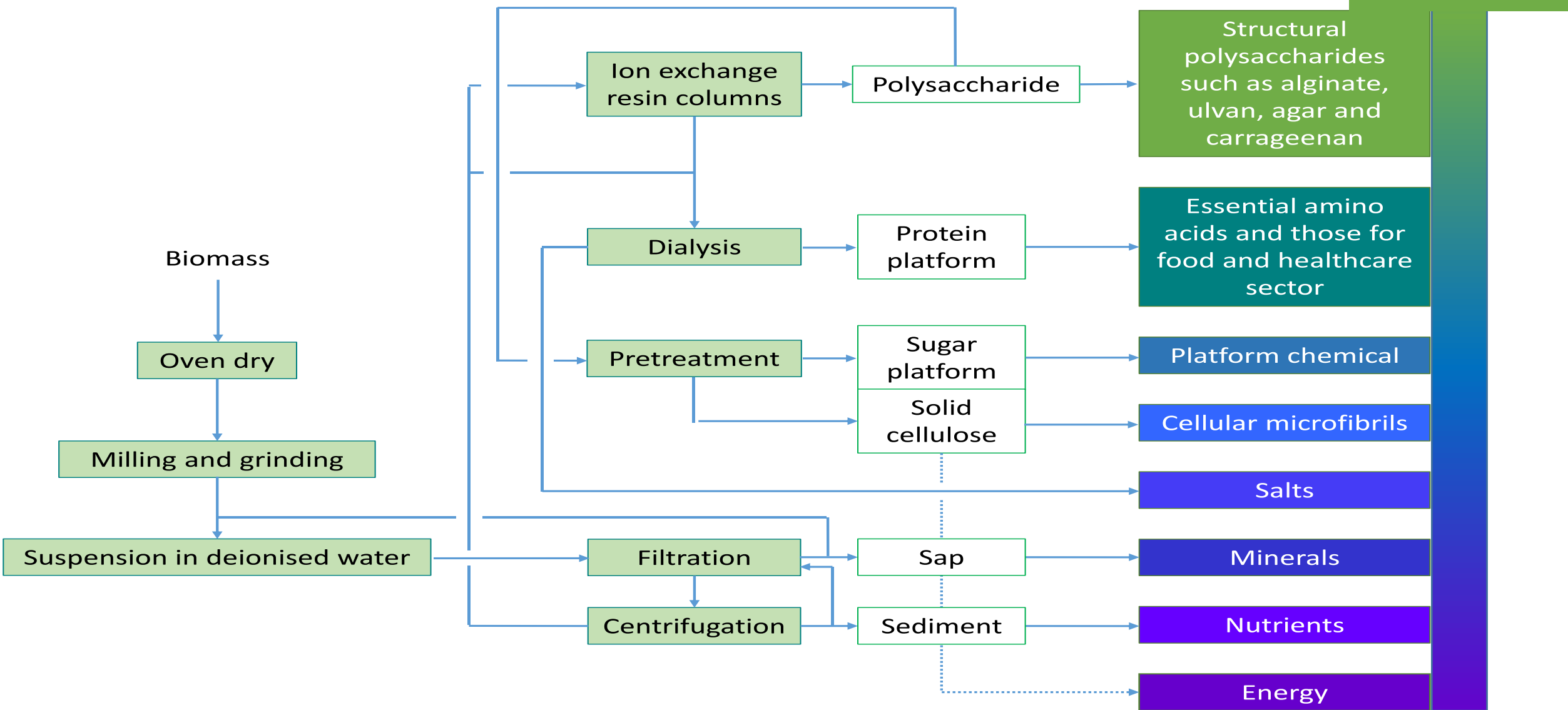
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High value low volume

Low value high volume



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# Protein

- Glutamic acid, aspartic acid and arginine in high concentrations (5-25% by mass of protein), glycine, proline, serine, alanine, valine, threonine, phenylalanine, lysine and leucine in medium concentrations (4-9%) and histidine, isoleucine, methionine and tyrosine in low concentrations (<5%) can be extracted from protein depending on macroalgae strain and protein extraction protocol.
- Amongst these, valine, threonine, phenylalanine, lysine, leucine, histidine, isoleucine and methionine have been classified as essential amino acids for human body.
- Glutamic acid salt is a food additive and flavour enhancer, aspartic acid is a building block for protein synthesis, and arginine helps in neurotransmission and blood flow, glycine in sleep and memory, proline in digestive health, serine in brain, muscle and skin health, and alanine in blood sugar level.
- Although currently lacking standard techniques for extraction of these amino acids, recognising their important roles in food and medicines, there will be further value generation propositions utilising the protein platform from cultivated macroalgae.

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# Polysaccharides

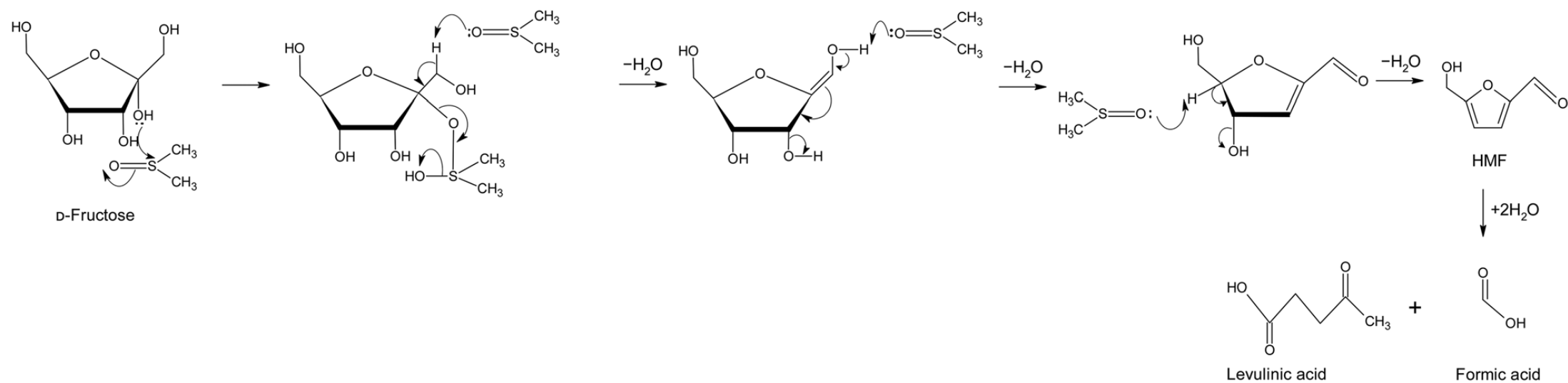
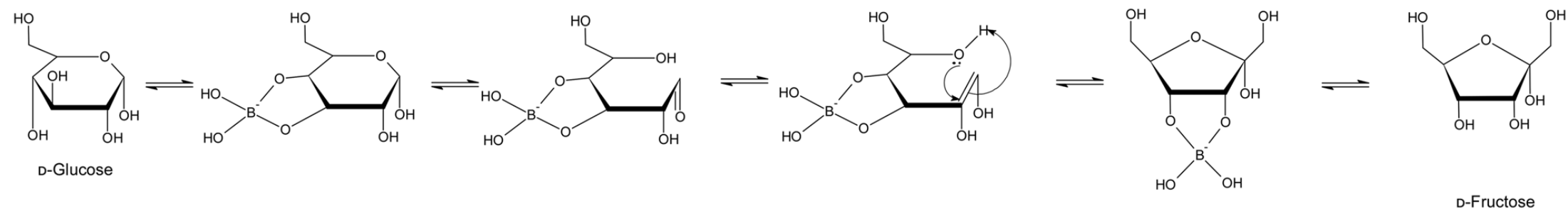
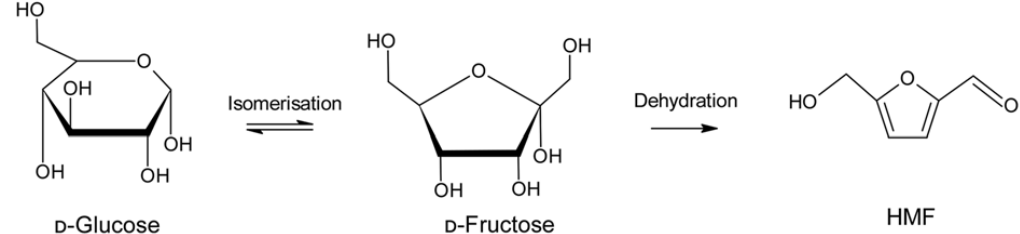
- The constituents of polysaccharides play two significant roles that also determine their application for human consumption.
- The two roles are energy storage and structural polysaccharides.
- The energy storing polysaccharides in brown, green and red algae are laminarin consisting of 20-25 glucose units, starch, and floridean starch and floridoside, with a structure similar to common starch.
- The structural polysaccharides in these macroalgae types include alginate; ulvan and cellulose; and cellulose, agar and carrageenan, respectively.

# Polysaccharides

- The laminarin structure may vary in degree of branching, the degree of polymerization and the ratio of (1,3)- and (1,6)-glycosidic bonds. Extracted from brown algae, they offer biological activities such as antioxidant, antitumor, antimicrobial, immune modulation, drug delivery and anticoagulant properties that determine their market applications such as functional foods and nutraceuticals and price as high as \$250 g<sup>-1</sup>.
- Starch consists of two types of molecules: the linear and helical amylose and the branched amylopectin. Depending on the plant, starch generally contains 20 to 25% amylose and 75 to 80% amylopectin by weight. Its main uses are as food conditioner, thickener and additive (~\$6 kg<sup>-1</sup>).
- Alginate is made of uronic acids: mannuronic and guluronic acids. Alginates are used to increase viscosity, to form gels and jellies and to stabilise food and in textiles by giving a smooth texture (\$10 kg<sup>-1</sup>).
- Ulvans are acidic water-soluble sulphated heteropolysaccharides that contribute to the strength of the cell wall and give flexibility to the plant. Their applications in the form of gels include chemical, pharmaceutical, biomedical and agricultural industries (\$395 g<sup>-1</sup>).
- Celluloses are polymers of C6 sugars linked by  $\beta$ -(1–4)-glycosidic bonds. Celluloses could be a feedstock to applications ranging from energy, biofuel, through chemical to food (\$4 kg<sup>-1</sup>).
- Agar is made of agarose and agarpectin, with agarose making up about 70% of the mixture. Agarose is a linear polymer, made up of repeating units of agarobiose, a disaccharide made up of D-galactose and 3,6-anhydro-L-galactopyranose. Agar can be used as a laxative, an appetite suppressant, a vegetarian substitute for gelatin, a thickener for soups, in fruit preserves, ice cream, and other desserts, as a clarifying agent in brewing, and for sizing paper and fabrics (\$40 g<sup>-1</sup>).
- *Carrageenan* is a linear polysaccharide made up of a repeating dissacharide sequence of  $\alpha$ -D-galactopyranose and  $\beta$ -D-galactopyranose. Carrageenan is used as a binder, thickening agent, and a stabiliser in medications, foods, and toothpaste. Carrageenan is also an ingredient in weight loss products (\$30 kg<sup>-1</sup>).

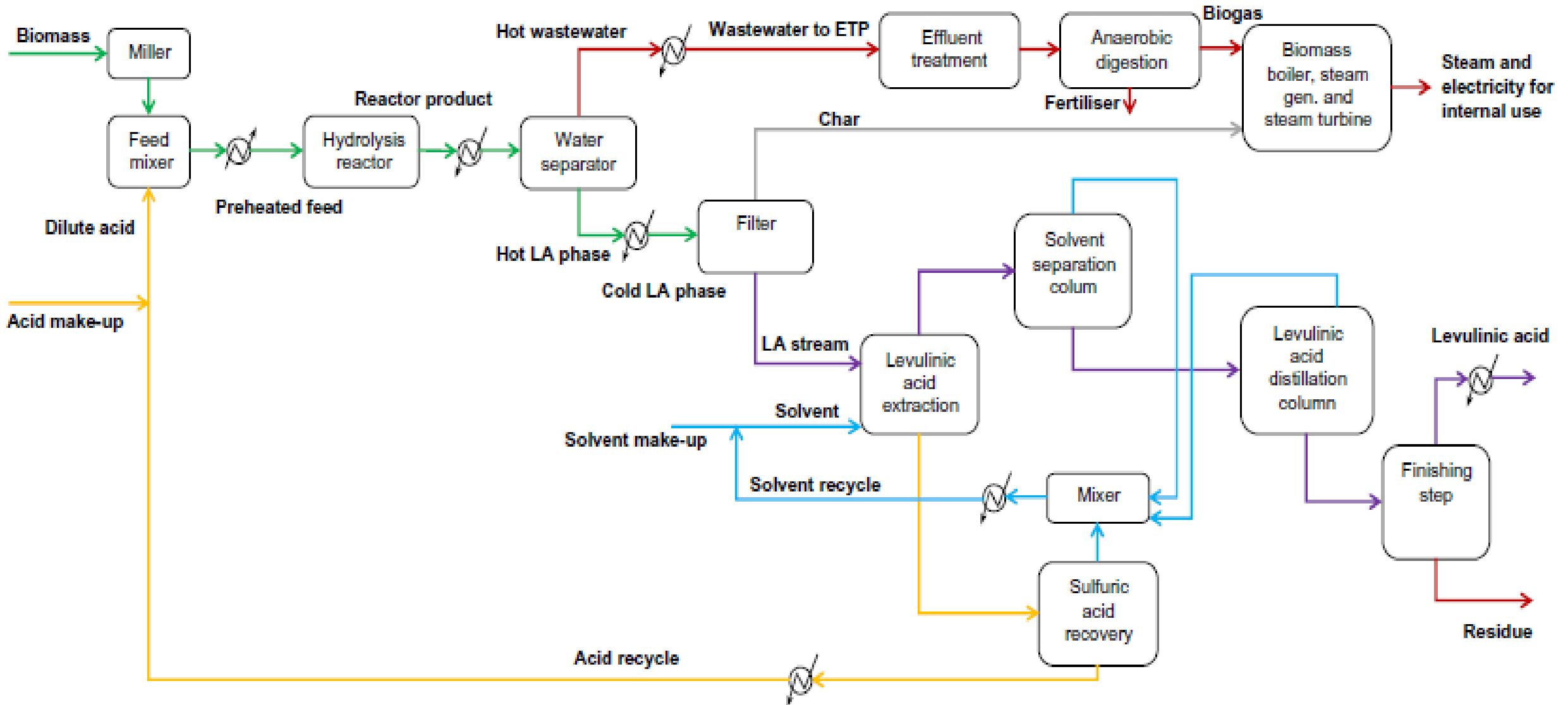
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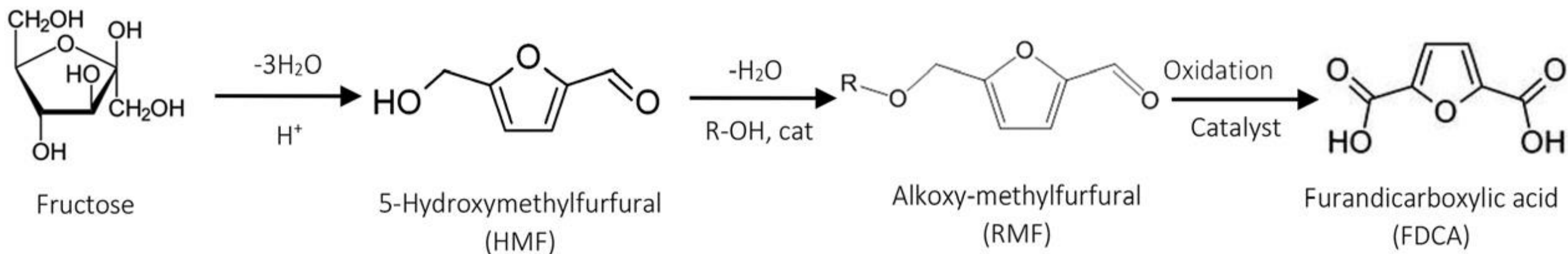


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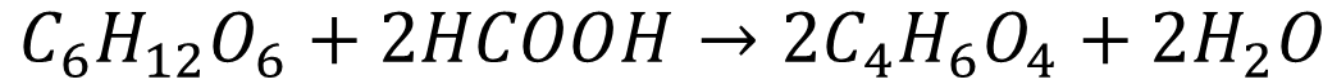
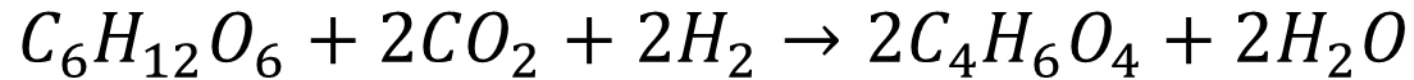
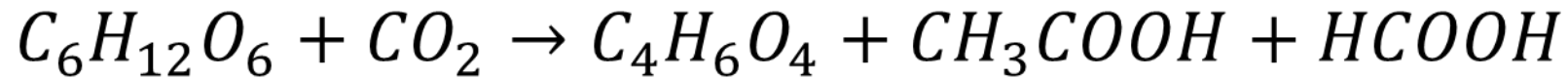
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# Production of succinic acid by CO<sub>2</sub> reuse



# Succinic Acid Production

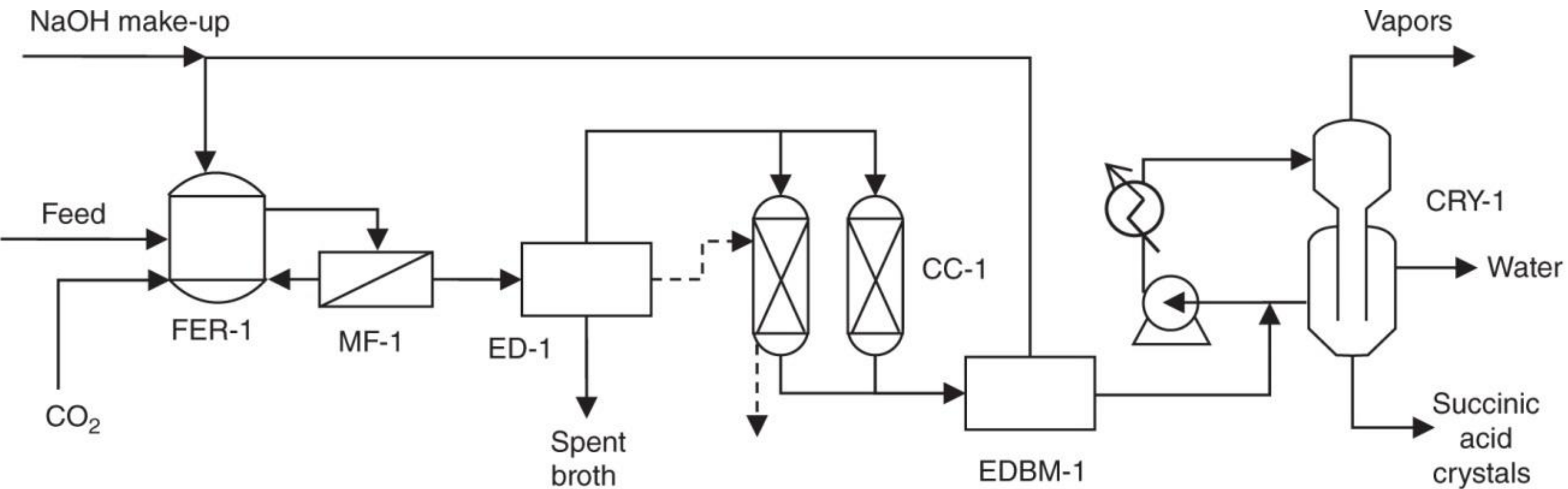
- Depending on the microorganism, fermentation technology and operating conditions, other metabolites such as ethanol, lactic acid, acetic acid and formic acid are formed as by-products.
- The viability for a commercial technology resides in applying metabolic engineering to favour the fermentative pathways that minimise by-product formation.
- A maximum theoretical yield of 2 moles succinic acid per mole glucose can be achieved, when using a mixture of  $\text{CO}_2$  and  $\text{H}_2$ .
- Carbon feedstock, pH, carbon dioxide and hydrogen are critical for the production of succinate. A proper combination of these parameters must be selected for each microorganism, as they use different pathways for succinate production and tolerate different levels of  $\text{CO}_2$ , pH and  $\text{H}_2$ .
- $\text{CO}_2$  is an electron acceptor that diverts metabolism to pyruvate and lactate/ethanol when present at low levels but to succinate when present at high levels.
- $\text{CO}_2$  can be supplied from an exhaust gas stream from post-combustion or combined heat and power generation, fermentation to bioethanol production, carbonates added to the medium (e.g.  $\text{MgCO}_3$ ,  $\text{Na}_2\text{CO}_3$ ,  $\text{NaHCO}_3$ , or  $\text{CaCO}_3$ ) or from a combination of these sources.

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**Figure 11.41** Succinic acid production process using simultaneous acidification and purification by electrodialysis and crystallization. FER: fermentor, MF: microfiltration, ED: desalting electrodialysis, EDBM: electrodialysis with bipolar membrane, CC: chelation columns, CRY: crystallizer. Dashed lines operate only during regeneration of the ion exchanger.

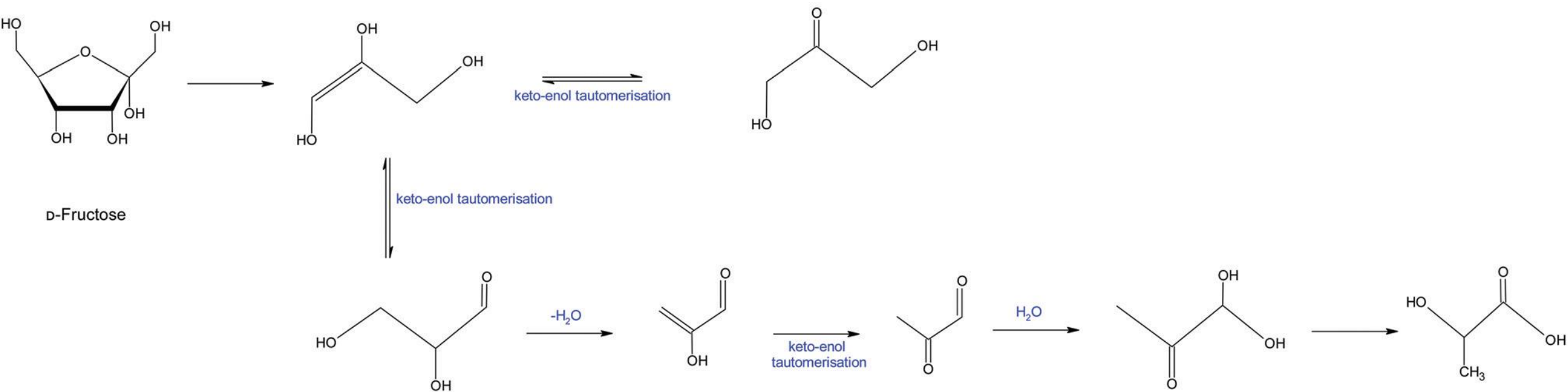
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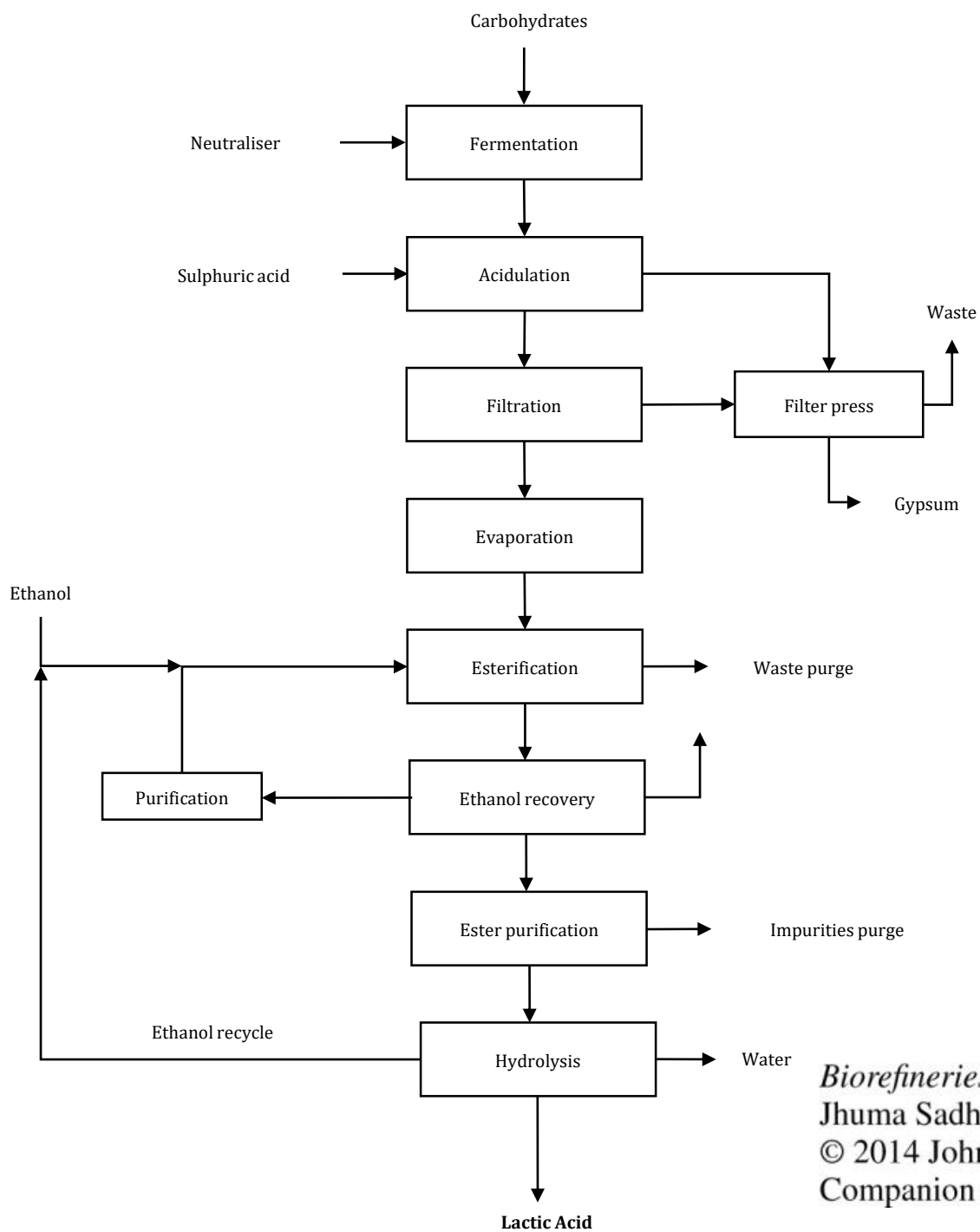
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**Fructose to Lactic acid**



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$$\text{Net Present Value} = \sum_{\text{plant life in years}} \frac{\text{Net Cash Flow}}{(1 + \text{Discount rate})^{\text{year}}}$$

$$= \sum_{\text{plant life in years}} \frac{(\text{Revenues} - \text{Annual capital cost} - \text{Total OPEX})}{(1 + \text{Discount rate})^{\text{year}}}$$

$$\text{Annual capital cost} = \text{Annual Capital Charge} \times \text{Total capital cost}$$

$$\text{Desired payback time} = f(\text{Annual Capital Charge}, \text{Discount rate})$$

Total capital cost = 5 × Delivered cost of equipment  
 Delivered cost of equipment:  
 Apply scaling factor for economy of scale and levelise the cost

$$\text{Total OPEX} = 1.3 \times (\text{fixed operating cost} + \text{variable operating cost})$$

$$\begin{aligned} \text{fixed operating cost} \\ &= 0.55 \times \text{Delivered cost of equipment} + 2 \\ &\times \text{personnel cost} \end{aligned}$$

$$\begin{aligned} \text{variable operating cost} = \\ \text{total cost of feedstock, utilities and reagents} \end{aligned}$$

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	Base cost million \$	Scale factor	Base capacity	Current capacity	Unit	CEPCI at base year	Current levelised delivered cost of equipment million \$	Recent CEPCI
Drying	7.6	0.8	33.5	1.14	t h <sup>-1</sup> wet biomass	394.3	0.73	567.5
Milling	0.37	0.7	50	0.33	t h <sup>-1</sup> biomass (30% moisture w/w)	402	0.02	567.5
Grinding	0.41	0.6	33.5	0.33	t h <sup>-1</sup> biomass (30% moisture w/w)	394.3	0.04	567.5
Filtration	2.92	0.7	18.5	0.23	t h <sup>-1</sup> deionised water	402	0.19	567.5
Centrifugation	1.05	0.65	10.1	0.01	t h <sup>-1</sup> dry solid	402	0.02	567.5
Ion exchange resin column	2.39	0.33	83.3	0.22	t h <sup>-1</sup> dry feed	402	0.47	567.5
Dialysis	1.05	0.65	10.1	0.03	t h <sup>-1</sup> dry solid	402	0.04	567.5
Pretreatment	1.41	0.78	83.3	0.16	t h <sup>-1</sup> dry feed	402	0.02	567.5
Integrated biorefinery producing protein, sugar and inorganic platforms							1.52	
Levulinic acid production from sugar platform							1.95	
FDCA production from sugar platform							1.22	
Succinic acid production from sugar platform (crystallisation)							3.99	
Lactic acid production from sugar platform							1.73	

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Processing step	Inventory
Oven dry	60-90% moisture content by mass of wet macroalgae; oven dry at 60°C for 2-3 days. 10 MJ heat.
Milling and grinding	0.05 kWh power
Suspension in deionised water	2 kg deionised water proportional to the mineral / nutrient / salt contents
Filtration	0.4 kWh power
Centrifugation	3 kWh power
Dialysis	0.075 kWh on the basis of salt removal
Pretreatment	0.5 kJ for polysaccharides decomposition into sugar platform. Could vary between 0.05-5 kJ depending on g mol <sup>-1</sup> of polysaccharides
Levulinic acid production from sugar platform	Yield: 0.46 (w levulinic acid / w cellulose). 0.5% (w make up sulphuric acid / w sugar). 4 MJ heat. Residual organic stream 8%
FDCA production from sugar platform	Yield: 0.64 (w FDCA / w sugar). Energy use for electricity, heat and reagent supplies: 5 MJ
Succinic acid production from sugar platform (crystalisation)	Yield: 0.75 (w succinic acid / w sugar). 0.5% (w make up hydrochloric acid / w sugar). 4.3 MJ heat and 0.9 kWh power
Lactic acid production from sugar platform	Yield: 0.93 (w lactic acid / w sugar). Energy use for electricity, heat and reagent supplies: 6 MJ

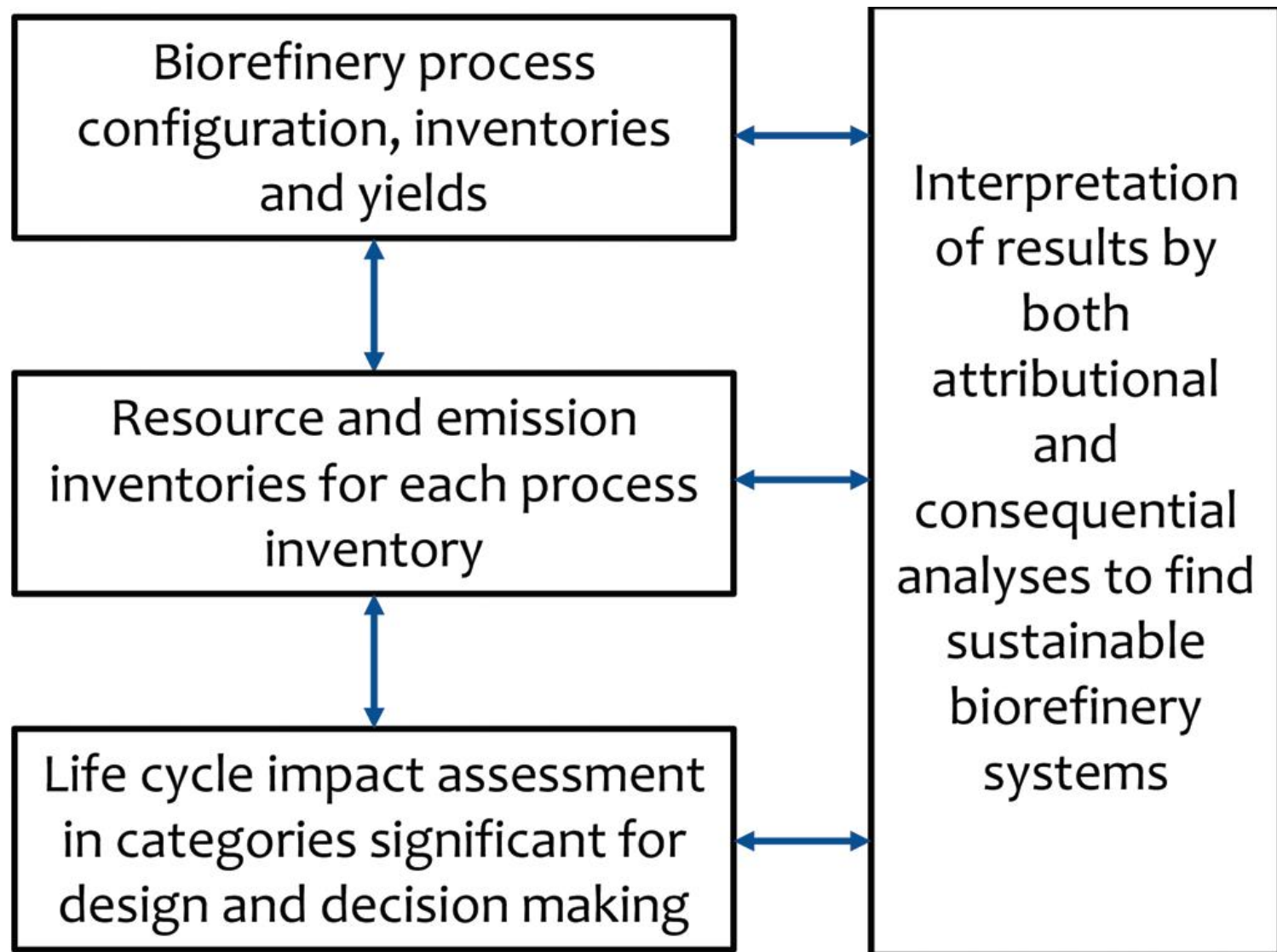
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# Technoeconomic assessment results

- Electricity and steam prices are \$0.046 and \$0.005 MJ<sup>-1</sup>, respectively.
- Based on a cost of \$50 t<sup>-1</sup> of dry macroalgae (0.23 tph), the annual cost of production from the integrated biorefinery system is 3.7 million \$ y<sup>-1</sup>.
- This gives the cost of production of each product from the system at \$2010 t<sup>-1</sup>, significantly lower than the market price of the polysaccharides, i.e. upto \$395 g<sup>-1</sup> (ulvans) as well as the market price of essential amino acids \$58 kg<sup>-1</sup>, making the integrated biorefinery system economically feasible.
- The cost of production is also lower than the market price of salts (\$7 kg<sup>-1</sup>), but higher than the market price of nutrients (\$5.4 t<sup>-1</sup>).
- As the production rate of nutrients is insignificant, an overall profitable economic performance of the macroalgae integrated biorefinery system is forecasted.

# Technoeconomic assessment results

- The prices (\$ t<sup>-1</sup>) of levulinic acid, FDCA, succinic acid and lactic acid are 4500, 2450, 1800 and 1300, respectively.
- Their yields (tph) from the sugar platform are 0.021, 0.1, 0.12 and 0.15, respectively.
- For the given cost of \$50 t<sup>-1</sup> of dry macroalgae, the annual cost of production (in million \$ y<sup>-1</sup>) of levulinic acid, FDCA, succinic acid and lactic acid from the sugar platform (without the cost of production of the sugar platform) is 2.7, 2.2, 2.2 and 3, respectively.
- However, when the cost of production of the sugar platform of \$2010 t<sup>-1</sup> is added, their annual cost of production (in million \$ y<sup>-1</sup>) becomes 3.7, 5.3, 4.8, 8.4 and 5.6, respectively.
- These costs of production in \$ t<sup>-1</sup> of the chemical product are equal to 2895, 2620, 4620 and 3055, respectively, indicating decreasing economic feasibility in the order of: levulinic acid > FDCA > lactic acid > succinic acid, respectively.
- When the chemical production from the sugar platform is combined with the protein extraction in an overall integrated biorefinery system, the production of any building block or platform or “sleeping giant” chemical utilising the sugar platform becomes economically feasible.
- Based on the revenues generated from the extracted protein and chemical alone, a discounted cash flow of 4-8 million \$ y<sup>-1</sup> (\$440-880 t<sup>-1</sup> wet seaweed) is estimated, with decreasing order of profitability from FDCA through lactic acid and levulinic acid to succinic acid, respectively.

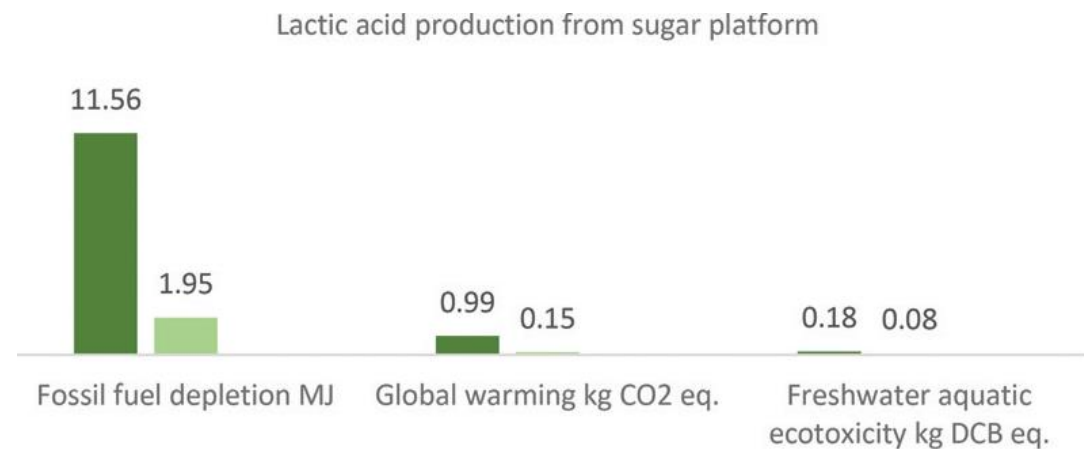
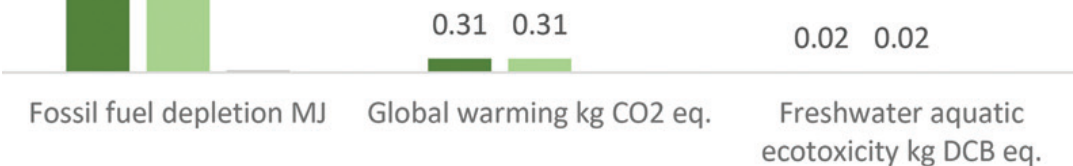
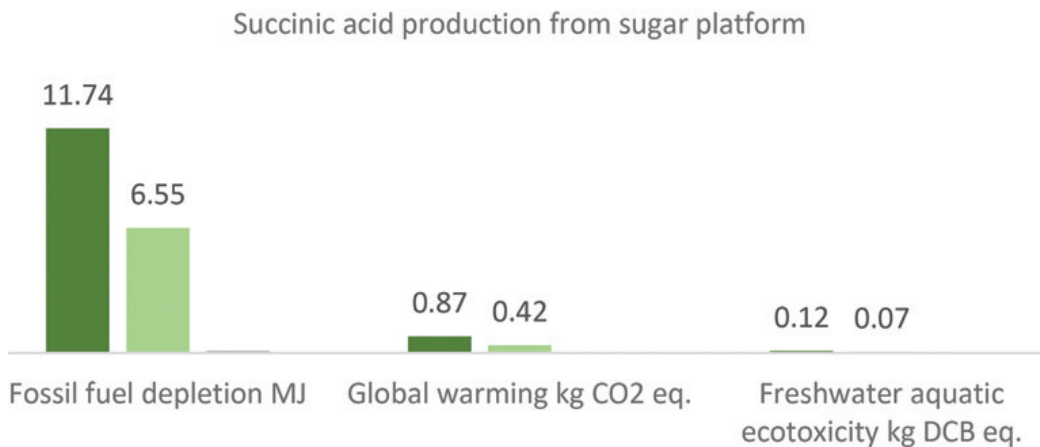
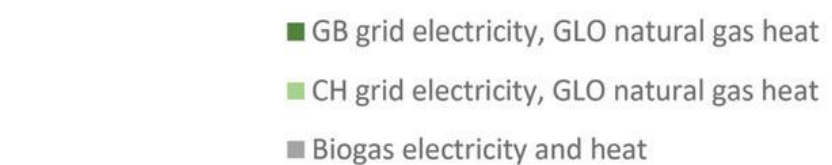
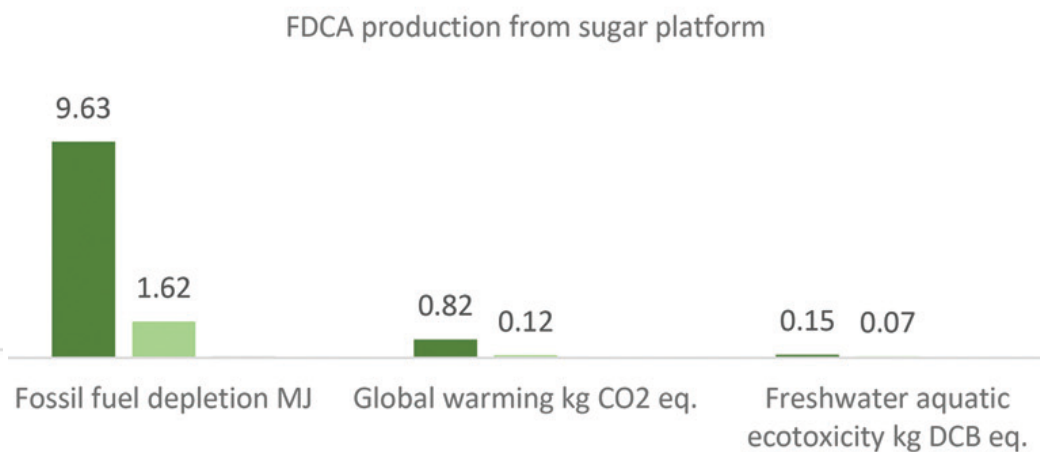
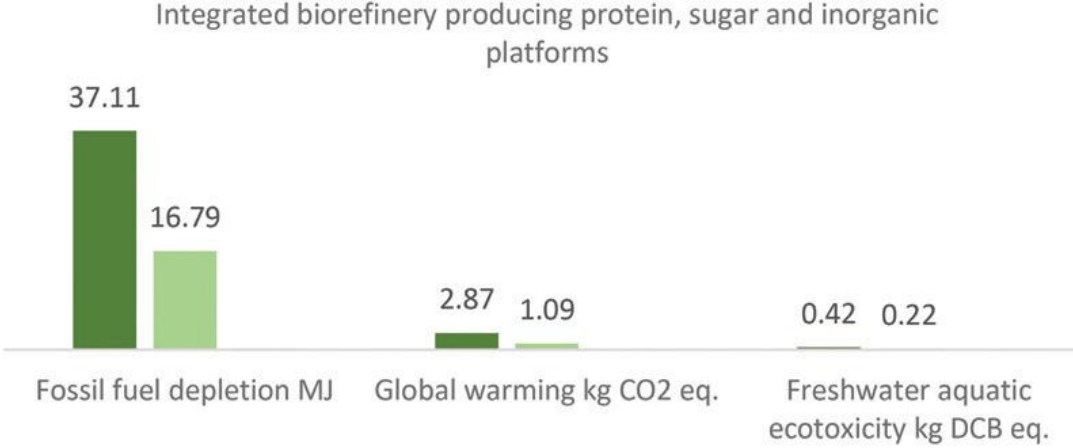


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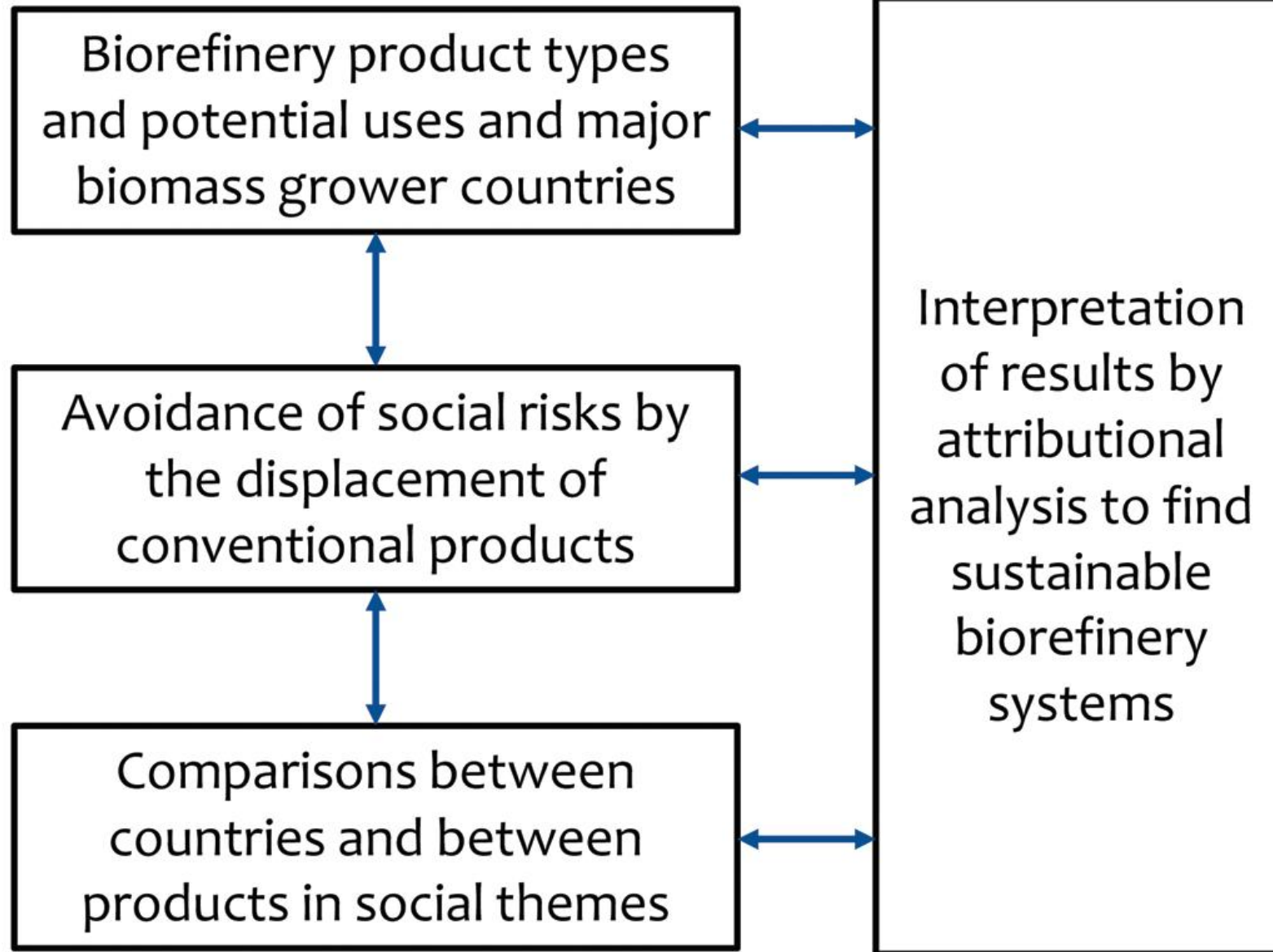
<b>Electricity</b> , high voltage { <b>CH</b> }  market for   Ecoinvent 3 - allocation, default - unit
<b>Electricity</b> , high voltage { <b>GB</b> }  production mix   Ecoinvent 3 - allocation, recycled content - unit
<b>Heat</b> , central or small-scale, natural gas { <b>GLO</b> }  market group for   Ecoinvent 3 - allocation, default – unit
<b>Biogas</b> { <b>GLO</b> }  market for   Ecoinvent 3 - allocation, default – unit
<b>Sulfuric acid</b> { <b>GLO</b> }  market for   Ecoinvent 3 - allocation, default – unit
<b>Hydrochloric acid</b> , without water, in 30% solution state { <b>RER</b> }  hydrochloric acid production, from the reaction of hydrogen with chlorine   Ecoinvent 3 - allocation, default - unit
<b>Lactic acid</b> { <b>RER</b> }  production   Alloc Def, U Ecoinvent 3 - allocation, default - unit

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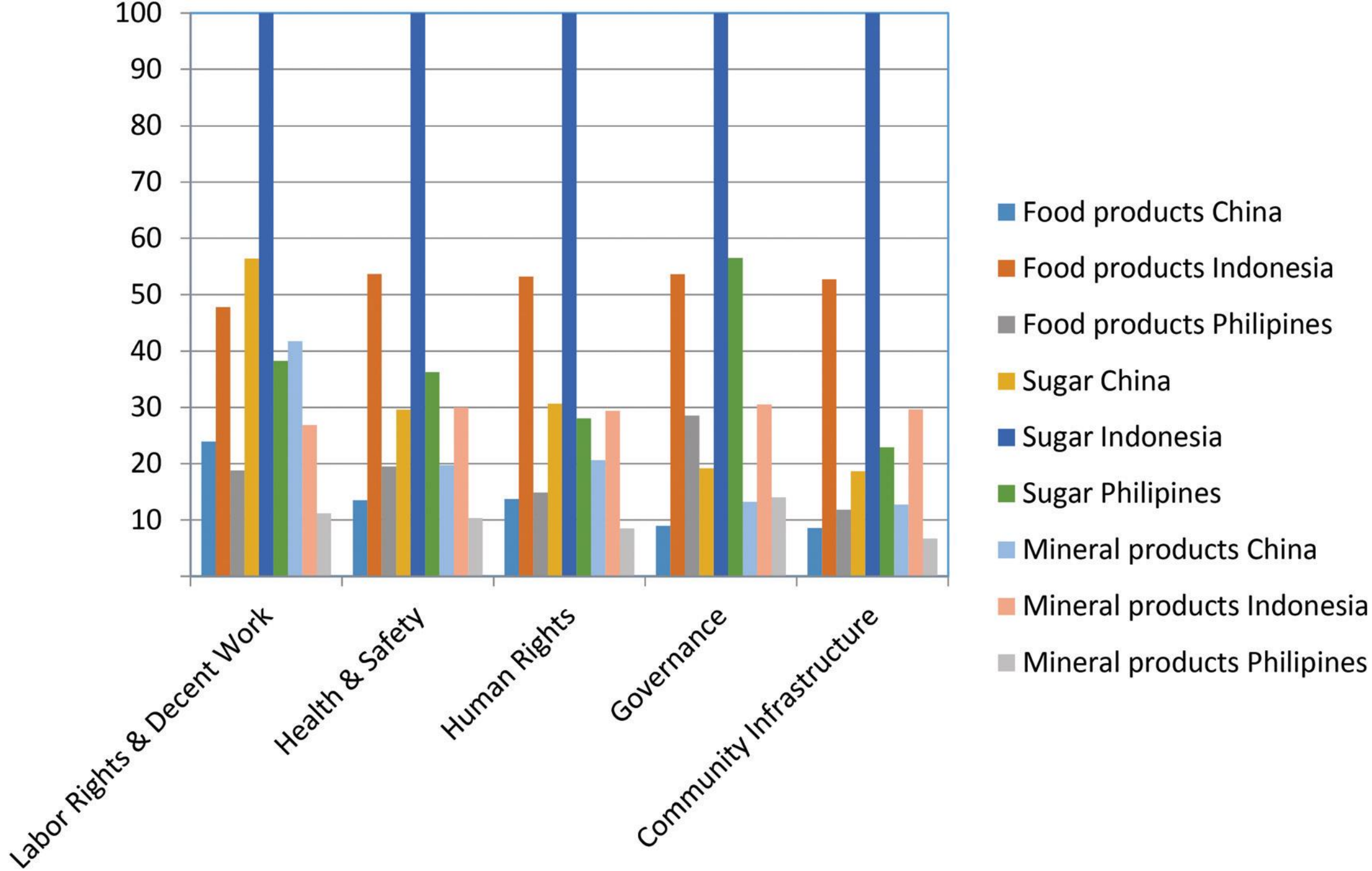


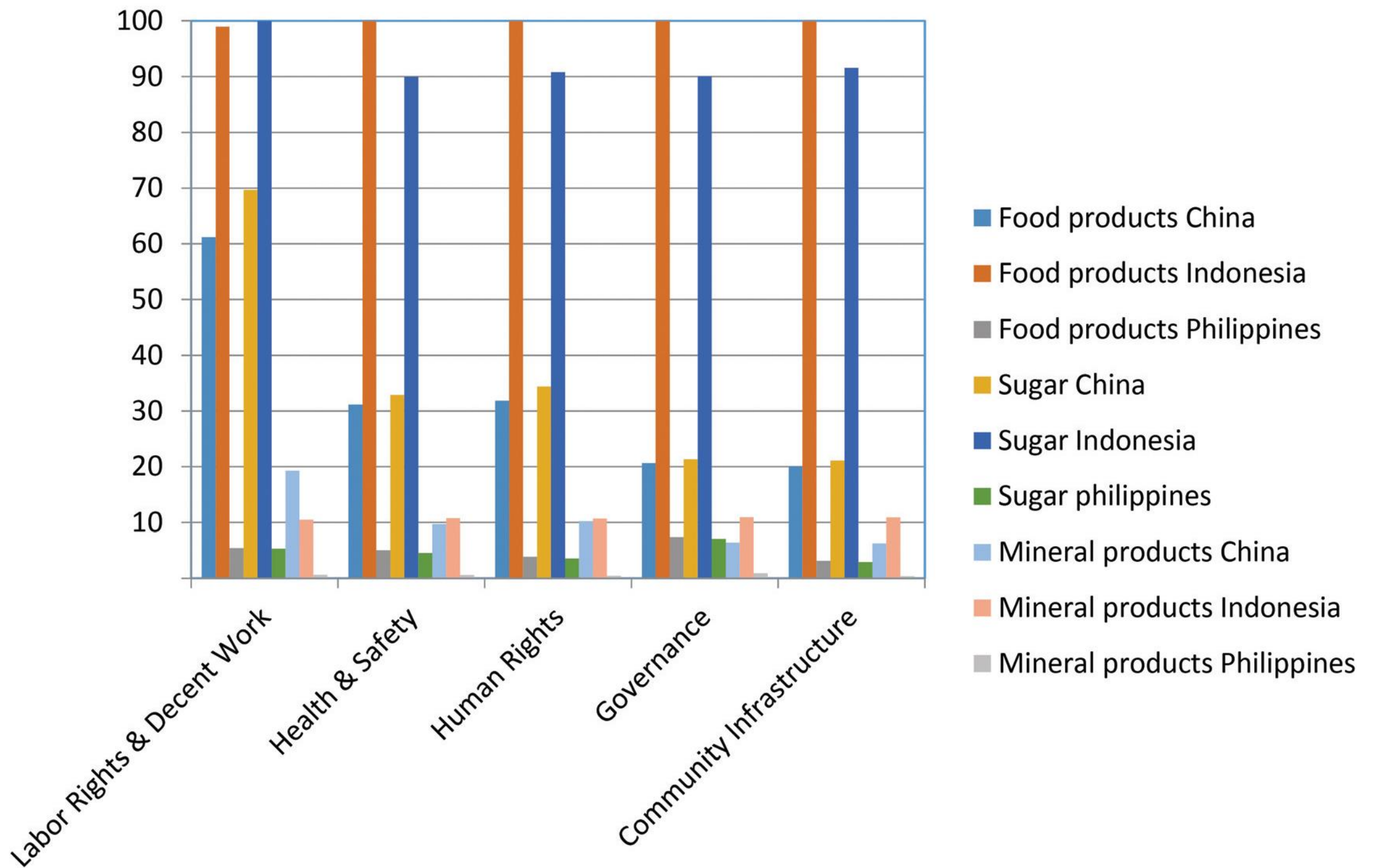
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# Social impact savings

- Social life cycle assessment (SLCA) estimates five main governing social categories, labour rights & decent work, health & safety, human rights, governance and community infrastructure.
- Among the different SLCA databases developed so far, the Social Hotspot Database (SHDB) is the most significant. Over a 5-years project, the New Earth involved stakeholders worldwide to assign importance and risk to each theme under each category in a sector of goods in a country. The supply chain interactions are considered in the computation of total index of a theme for a given sector in a given country. In constructing the SHDB, the team combined the Global Trade Analysis Project (GTAP) data on import-export of goods between countries and social risk characterisation data (from stakeholders) to calculate social attributional risks of products in two ways, countries of origin and life cycle approaches.
- The unit of risks is medium-risk hour (mrh) for a given functional unit, e.g. rate of product formation in case of a production system, in terms of its monetary value worth.
- The individual social theme risks are factored by the netted fractional imports considering the entire supply chain and these factored individual social theme risks are added to give a total social theme risk for given sector in given country to create a base case.
- Using the UNCOMTRADE data and the SHDB, a base case scenario and a scenario with 100% self-generation case (or no import) are compared in terms of weighted aggregated risks in social categories for a country, in the TESARREC platform.

*Biorefineries and Chemical Processes: Design, Integration and Sustainability Analysis*, First Edition.

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Companion Website: <http://www.wiley.com/go/sadhukhan/biorefineries>

# SLCA results for chemical product : Mexico case

Spain	23.57	21.72	3.87	5.70	2.86
USA	36.18	8.57	4.52	5.75	2.91
Netherlands	8.55	4.94	2.03	1.86	1.23
China	327.56	83.40	71.69	32.02	30.33
France	3.22	3.17	0.75	0.95	0.95
Belgium	10.08	4.57	2.44	3.17	2.65
Brazil	5.21	5.61	1.67	0.99	0.70
Germany	0.0096	0.0079	0.0022	0.0021	0.0026
Malaysia	1.35	0.62	0.27	0.34	0.07
Japan	0.004353	0.002250	0.000790	0.000924	0.000898
Colombia	0.000984	0.000415	0.000248	0.000373	0.000057

Mexico	311.19	87.23	89.14	129.75	12.84
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# Conclusions

- Novel biorefinery systems have been developed to coproduce sugar, protein, inorganic (salts, minerals and metals) and nutrient platforms from macroalgae by holistic rigorous systematic process modelling, synthesis and integration analyses applying green chemistry principles.
- The design activity provides the process inventories as the basis for the estimation of the capital and operation costs, costs of production of individual products, discounted cash flows, (environmental) life cycle assessments of overall systems as well as individual products and avoided environmental and social impacts due to displacement of individual fossil derived products.
- Important insights have been generated into target products from synthesis, integration and life cycle sustainability assessments comprising economic, environmental and social aspects.
- Sustainability of sugar derived platform chemical strongly depends on how energy is resourced. With increasing self-generation in terms of on-site combined heat and power or bioenergy generation and decreasing fossil based external energy supply, feasibility of sugar derived platform chemical increases.

Sadhukhan, J., Gadkari, S., Martinez-Hernandez, E., Ng, K.S., Shemfe, M., Torres-Garcia, E. and Lynch, J., 2019. Novel macroalgae (seaweed) biorefinery systems for integrated chemical, protein, salt, nutrient and mineral extractions and environmental protection by green synthesis and life cycle sustainability assessments. *Green Chemistry*, 21(10), pp.2635-2655. <https://doi.org/10.1039/C9GC00607A>