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Review

Recent advances in pretreatment of lignocellulosic wastes and production of value added products

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This study highlights the recent advances in the treatment and value addition of lignocellulosic wastes (LCW) with main focus on domestic and agro-industrial residues. Mechanical, physical and biological treatment systems are brought into perspective. The main value-added products from lignocellulosic wastes are summarized in a manner that pinpoints the most recent trends and the future directions. Physicochemical and biological treatment systems seem to be the most favored options while biofuels, biodegradable composites and biosorbents production paints a bright picture of the current and future bio-based products. Engineered microbes seem to tackle the problem of bioconversion of substrates that are otherwise non convertible by conventional wild strains. Although the main challenge facing LCW utilization is the high costs involved in treatment and production processes, some recent affordable processes with promising results have been proposed. Future trends are being directed to nanobiotechnology and genetic engineering for improved processes and products. The paper presents state of the art review of the dual advantage of handling LCW for cleaner environment and production of renewable bio-products.

Key words: Lignocellulosic wastes, pretreatment systems, value-added products.

INTRODUCTION

Lignocelluloses wastes (LCW) refer to plant biomass wastes that are composed of cellulose, hemicellulose, and lignin. They may be grouped into different categories such as wood residues (including sawdust and paper mill discards), grasses, waste paper, agricultural residues (including straw, stover, peelings, cobs, stalks, nutshells, non food seeds, bagasse, domestic wastes (lignocellulose garbage and sewage), food industry residues, municipal solid wastes and the like (Qi et al., 2005; Roig et al., 2006; Rodríguez et al., 2008). Currently, the second generation bio-products such as bioethanol, biodiesel, biohydrogen and methane from lignocellulose biomass are increasingly been produced from wastes rather than from energy crops (jatropha, switchgrass, hybrid poplar and willow) because the latter competes for land and water with food crops that are already in high demand. The use of food crops such as corn and sugarcane to produce biofuels is increasingly being discouraged due to the current worldwide rise in food prices. In order to minimize food-feed-fuel conflicts, it is necessary to integrate all kinds of biowaste into a biomass economy (Mahro and Timm, 2007). Furthermore, the use of LCW offers a possibility of geographically distributed and greenhouse-gas-favourable sources of products (Rubin, 2008).

The lignocellulosic biomass, which represent the largest renewable reservoir of potentially fermentable carbohydrates on earth (Mtui and Nakamura, 2005), is mostly wasted in the form of pre-harvest and post-harvest agricultural losses and wastes of food processing industries. Due to their abundance and renewability, there has been a great deal of interest in utilizing LCW for the production and recovery of many value-added products (Pandey et al., 2000; Das and Singh, 2004; Foyle et al., 2007). Among the main recovery products include enzymes, reducing sugars, furfural, ethanol, protein and amino acids, carbohydrates, lipids, organic acids, phenols, activated carbon, degradable plastic composites, cosmetics, biosorbent, resins, medicines, foods and feeds, methane, biopesticides, biopromoters, secondary metabolites, surfactants, fertilizer and other miscellaneous products (Tengerdy and Szakacs, 2003; Mtui, 2007; Ubalua, 2007; Galbe and Zacchi, 2007; Demirbas, 2008). Alongside producing these products, the processes also remove wastes from the environment.

The barrier to the production and recovery of valuable materials from LCW is the structure of lignocellulose which has evolved to resist degradation due to cross-linking between the polysaccharides (cellulose and hemicellulose) and the lignin via ester and ether linkages (Yan and Shuya, 2006; Xiao et al., 2007). Cellulose, hemicellulose and lignin form structures called microfibrils, which are organized into microfibrils that mediate structural stability in the plant cell (Rubin, 2008). The main goal of any pretreatment, therefore, is to alter or remove structural and compositional impediments to hydrolysis and subse-quent degradation processes in order to enhance digestibility, improve the rate of enzyme hydrolysis and increase yields of intended products (Mosier et al., 2005; Hendriks and Zeeman, 2009). These methods cause mechanical, physical chemical or biological changes in the plant biomass in order to achieve the desired products.

Technology of LCW bioconversion has long been considered to be rather expensive. However, recent increases in grain prices mean that the switch to second generation bio-products such as biofuels from LCW will reduce competition with grain for food and feed, and allow the utilization of materials like straw which would otherwise go to waste. Technologies that will allow cost-effective conversion of biomass into fuels and chemicals consider economy of scale, low-cost pretreatment systems and highly effective and efficient biocatalysts (Schneider and McCar, 2003; Gray et al., 2006).

This work reviews the recent developments in LCW pretreatment, value addition and techno-economic considerations.

PRETREATMENT TECHNOLOGIES FOR LIGNOCELLULOSIC WASTES

Mechanical pretreatment

Mechanically based pretreatment technologies are aimed at reducing the size of LCW to facilitate subsequent treatments. Reduction of biomass size below #20 sieves shows the best mechanical performance (de Sousa et al., 2004). Mechanical pretreatment technologies increase the digestibility of cellulose and hemicellulose in the lignocellulosic biomass. The use of mechanical chopping (de Sousa et al., 2004); hammer milling (Iñiguez-Covarrubias et al., 2001; Mani et al., 2004); grind milling (Mtui and Nakamura, 2005); roll milling (Qi et al., 2005); vibratory milling (Guerra et al., 2006) and ball milling (Inoue et al., 2008) have proved success as a low cost pretreatment strategy. The pulverized materials with increased surface area have been found to facilitate the subsequent physicochemical and biochemical pretreatments of corn stover, barley straw sugar cane baggase, wheat straw, wood waste and municipal solid waste. They result to improved digestibility of cellulose and hemicellulose to glucan and xylan, respectively; they further enhance enzymatic digestibility with lower enzyme loads. Mechanical pretreatment also result to substantial lignin depolymerization via the cleavage of uncondensed-aryl ether linkages (Inoeu et al., 2008). Solubility and fermentation efficiency of the natural lignocellulosic residues is also substantially increased by mechanophysicochemical pretreatment, leading to value-added utilization of these residues (Qi et al., 2005).

Physical pretreatment

Elevated temperatures and irradiation are the most successful physical treatments in the processing of LCW. Thermogravimetric treatment of wood waste under both inert and oxidant atmospheres from room temperature up to 1100 K leads to moisture loss; hemicellulose, cellulose and lignin decomposition (Lapuerta et al., 2004). On the other hand, pyrolysis of nutshells, straws, sawdust and municipal solid wastes at temperatures of 600 - 1200 K result to yields of char, liquid and gaseous products of up to 55% of the original LSW (Puértolas et al., 2001; Demirbas, 2002; Bonelli, 2003; Chen et al., 2003; Álvarez et al., 2005; Phan et al., 2008; Zabaniotou et al., 2008).

Irradiation can cause significant breakdown of the structure of LSW. Microwave irradiation at a power of up to 700 W at various exposure times resulted to weight loss due to degradation of cellulose, hemicellulose and lignin, and the degradation rates are significantly enhanced by the presence of alkali (Zhu et al., 2005a, 2005b, 2006). In addition, gamma radiation has been shown by Yang et al. (2008) to cause significant breakdown of the structure of powder of 140 mesh wheat straw, leading to weight loss and glucose yield of 13.40% at 500 kGy.

Physicochemical pretreatment

Combined chemical and physical treatment systems are of importance in dissolving hemicellulose and alteration of lignin structure, providing an improved accessibility of the cellulose for hydrolytic enzymes (Hendriks and Zeeman, 2009). The most successful physicochemical preatments include thermochemical treatments such as steam explosion or (steam disruption), liquid hot water (LHW), ammonia fiber explosion (AFEX) and CO_2 explosion (Sun and Cheng, 2002). In these processes, chipped biomass is treated with high-pressure saturated steam, liquid ammonia or CO_2 and then the pressure is swiftly reduced, making the materials to undergo an explosive decompression.

Steam explosion is typically initiated at a temperature of 160 – 260°C (corresponding pressure of 0.69 – 4.83 MPa) for several seconds to a few minutes before the material is exposed to atmospheric pressure. The processes cause hemicellulose degradation and lignin transformation due to high temperature, thus increasing the potential of cellulose hydrolysis. Addition of H_2SO_4 (or SO_2) or CO_2 in steam explosion of LCW can effectively improve enzymatic hydrolysis, decrease the production of inhibitory compounds, and lead to more complete liquefaction of hemicellulose, glucan, xylan, mannan, galactan, and arabinan (Jeoh and Agblevor, 2001; Sun and Cheng, 2002). Such pretreatments also lead to higher digestion efficiencies during production of monosaccharides, oligosaccharides, lactic acid, antibacterial violet pigments and methane gas (Liu et al., 2002; Kim et al., 2003; Asada et al., 2005; Wang and Chen, 2007; Öhgren et al., 2007). Wet oxidation pretreatment at 200 - 210°C in the presence of alkali or Na_2CO_3 leads to LCW solubilization and better enzymatic convertibility to value-added products (Fox and Noike, 2004; Lissens et al., 2004; Martín et al., 2008).

Liquid hot water (LHW) pretreatment utilizes pressurized hot water at pressure less than 5 Mpa and temperature range of 170 - 230°C for several minutes followed by decompression up to atmospheric pressure. Bagasse, corn stalk and straws of wheat, rice and barley pretreated by LHW have been reported to effect 80 - 100% hemicellulose hydrolysis, resulting to 45 - 65% xylose (Sun and Cheng, 2002; Sánchez and Cardona, 2008).

On the other hand, in AFEX treatment, the dosage of liquid ammonia ranging from 1 – 2 kg ammonia/kg dry biomass, temperature 90°C, and residence time of 30 min can significantly improve the saccharification rates (Chundawat et al., 2007; Thomsen and Belinda, 2007). On CO_2 explosion, 75% of the theoretical glucose released during 24 h of the enzymatic hydrolysis has been reported (Sun and Cheng, 2002). Ethanol yield of up to 83% of the theoretical value has been achieved for LCW subjected to physicochemical treatment (Jeoh and Agblevor, 2001).

Chemical pretreatment

Chemicals ranging from oxidizing agents, alkali, acids and salts can be used to degrade lignin, hemicellulose and cellulose from LCW. Poweful oxidizing agents such as ozone and H_2O_2 effectively remove lignin; does not produce toxic residues for the downstream processes; and the reactions are carried out at room temperature and pressure (Sun and Cheng, 2002). Alkali (NaOH, $Ca(OH)_2$, NaOH-urea, Na_2CO_3) hydrolyses of rice straw (Carrillo et al., 2005); spruce wood waste (Zhao et al., 2007); sugarcane, cassava and peanuts wastes (Thomsen and Belinda, 2007); corn cob (Torre et al., 2008); organic fraction of municipal solid waste (Torres and Lloréns, 2008) have been investigated. When these pretreatments are performed by using 0.5 - 2 M alkali at 120 - 200°C, they substantially facilitate saccharification and improve enzymic hydrolysis of LCW.

Dilute and concentrated acids at high temperature are suited for hydrolysis of LCW. Studies by del Campo et al. (2006) and Karimi et al. (2006) have established that 0.5% H_2SO_4 is optimal for treatment of wastes from vegetables and rice straw, respectively. More concentrated H_2SO_4 (up to 2.5 M) has been shown to be able not only to hydrolyse cellulose and hemicellulose, but also in separating lignin and other organic components from LCW (Iranmahboo et al., 2002; Alma and Acemioglu, 2004; Okafoagu and Nzelibe, 2006; Miller et al., 2007; Rahmanet al., 2007). SO_2 and fly ash in flare gas; HNO_3, HCl and polyhydric alcohol in the presence of sulfuric acid are also useful in LCW pretreatment (Fan, 2003; Herrera et al., 2004; Kobayashi et al., 2004; Rodríguez-Chonga et al., 2004; Hassan and Shukry, 2008): Recent studies have shown that when acids are combined with alkali, they play a more effective role in LCW pretreatment than acids and alkalis alone (Damisa et al., 2008).

Organic acids such as oxalic, acetylsalicylic and salicylic acid can be used as catalysts in the *organosolv* process whereby an organic or aqueous organic solvent mixture with inorganic acids (HCl or H_2SO_4) are used to break the internal lignin and hemicellulose bonds. The organic solvents used in the process include methanol, ethanol, acetone, ethylene glycol, triethylene glycol and tetrahydrofurfuryl alcohol (Sun and Cheng, 2002). The use of a dicarboxylic acid catalyst, maleic acid, for hemicellulose hydrolysis in corn stover overcomes the technical and economic hurdle of hemicellulose hydrolysis (Lu and Mosier, 2007).

Biological pretreatment

Biological treatment involves the use of whole organisms or enzymes in pretreatment of LCW. Both fungi and bacteria are used for biotreatment of LCW. Commercial preparations of fungal and bacterial hydrolytic and oxidative enzymes are also widely used instead of these microorganisms.

Fungal pretreatment of agricultural residues is a new method for improvement of digestibility (Sinegani et al., 2005). White-, brown- and soft-rot fungi are used to degrade lignin and hemicellulose in waste materials whereby brown rots mainly attack cellulose, while white and soft rots attack both cellulose and lignin. White-rot fungi are the most effective basidiomycetes for biological pretreatment of lignocellulosic materials (Sun and Cheng, 2002). Recent studies have shown that *Aspergillus terreus* (Emtiazi et al., 2001); *Trichoderma* spp (Pérez et al., 2002); *Cyathus stercoreus* (Keller et al., 2003); *Lentinus squarrosulus* (Shide et al., 2004); *Lentinus edodes* (Songulashvili et al., 2005; Brienzo et al., 2007); *Trametes pubes*cens (Melamane et al., 2007); *Pleurotus* spp (Ragunathan and Swaminathan, 2004; Mukherjee and Nandi, 2004; Belewu, 2006; Locci et al., 2008); *Penicillium camemberti* (Taşeli, 2008), *Phanerochaete chrysosporium* (Das and Hossain, 2000; Shi et al., 2008) grown at 25 - 35°C for 3 - 22 days resulted to 45 - 75% and 65 - 80% holocellulose and lignin degradation, respectively. The postreatement by anaerobic bioprocesses of LCW effluents that have been pretreated with fungi can lead to higher biogas than the original effluents (Coulibaly et al., 2003). Recombinant strains of *Saccharomyces cerevisiae* have been genetically engineered to carry out simultaneous saccharification and fermentation (SSF) to produce extracellular endo-glucanase and *β*-glucosidase that are able to ferment cellulose and hemicellulose to 6-carbon and 5-carbon sugars and subsequent fermentation to ethanol (Sedlak and Ho, 2004; van Maris et al., 2006; Haan et al., 2007; Chu and Lee, 2007; Wisselink et al., 2007). In bio-organosolv process, fungal (*Ceriporiopsis subvermispora*) pretreatment of wood waste for 2 - 8 weeks followed by organic solvent treatment at 140 - 200℃ for 2 h has achieved considerable energy efficient delignification and hemicellulose hydrolysis (Itoh et al., 2003; Sánchez and Cardona, 2008).

Bacterial pretreatment of LCW involves both anaerobic and aerobic systems. Anaerobic degradation utilizes mainly mesophillic, rumen derived bacteria (Han and Shin, 2002; Hu and Yu, 2005, 2006; Neves et al., 2006; Hu et al., 2008; Yue et al., 2008). Aerobic-anaerobic systems have an upper hand when it comes to degradation of LCW richer in lignin content (Ammary, 2004; Mshandete et al., 2005, 2008) while in aerobic system alone, actinomycete *Streptomyces griseus* is able to produce high levels of extracellular hydrolytic enzyme that degrade lignocellulose (Arora et al., 2005). *Escherichia coli* and *Klebsiella oxytoca* strains have been genetically engineered to produce microbial biocatalysts that produce bioethanol from lignocellulosic materials (Jarboe et al., 2007; Peterson and Ingram, 2008).

Enzymatic pretreatment of LCW utilize hydrolytic and oxidative enzymes which are mainly derived from fungi and bacteria. Cellulases are usually a mixture of several enzymes. At least three major groups of cellulases are involved in the hydrolysis process: (1) endoglucanase (endo-1,4-glucanohydrolase) which attacks regions of low crystallinity in the cellulose fiber, creating free chain-ends; (2) exoglucanase or cellobiohydrolase (CBH) (1,4-*β*-glucan cellobiohydrolase) which degrades the molecule further by removing cellobiose units from the free chain-ends and (3) *β*-glucosidase which hydrolyzes cellobiose to produce glucose (Sun and Cheng, 2002). In addition, there are also a number of ancillary enzymes that attack hemicellulose, such as glucuronidase, acetylesterase, feruloylesterase, xylanase, *β*-xylosidase, galactomanna-nase and glucomannanase (Nikolov et al., 2000; Draude et al., 2001; Aranda et al., 2004; Mtui and Nakamura, 2005, Roman et al., 2006; Georgieva et al., 2008). During the enzymatic hydrolysis, cellulose is degraded by cellulases to reducing sugars that can be fermented by yeasts or bacteria to ethanol.

Ligninolytic enzymes are primarily involved in lignin degradation in oxidative reactions that are mainly free radical driven in the presence (or sometimes absence) of mediators. The main enzymes involved are lignin peroxidase, manganese peroxidase and laccase (Hao et al., 2006; Mtui and Nakamura, 2007, 2008; Mtui and Masalu, 2008). The hydrolytic and oxidative enzymatic reactions are mainly carried out at 30 - 45℃ with low enzyme loading rate at reaction time of 6 - 26 h. All the pretreatment methods discussed above are summarized in Figure 1.

VALUE-ADDED PRODUCTS FROM LIGNOCELLULOSIC WASTES

Advances in industrial biotechnology offer potential opportunities for economic utilization of agro-industrial residues. Biodevelopment of biowastes provide a wide range of affordable renewable value-added products from LCW (Pandey et al., 2000; van Wyk, 2001; Howard et al., 2003).

Reducing sugars

Fermentable sugars comes first in the value chain of processed LCW with glucose, xylose, xylitol, cellobiose, arabinose, pentose and galactose being the main reduced sugars produced (Akmar and Kennedy, 2001; Saha, 2003; Rodríguez-Chonga et al., 2004; Yáñez et al., 2004; Sepúlveda-Huerta et al., 2006; Tabka et al., 2006; Hanchar et al., 2007; Singh et al., 2008; Li et al., 2008; Kim et al., 2008). In these sugar producing processes, hydrolysable sugars yield of up to 83.3% has been achieved at the reaction temperatures of 37 - 50℃ for 6 – 179 h at pH 5 - 6. The size of substrate added determines the amount of the saccharification products (Baig et al., 2004). In the enzymatic hydrolysis step using celluclast® supplemented with novozym®, a degree of saccharification of 100% has been achieved (Marques et al., 2008). Some transgenic plant residues have been reported to yield nearly twice as much sugar from cell walls compared to wild-types (Chen and Dixon, 2007). Glucose seems to be the major monosaccharide product from LCW. The challenge facing depolymerization of hemicellulose into fermentable sugars is the requirement for a consortium of enzymes to complete the hemi-cellulose hydrolysis, leading to high enzyme costs. Efforts to overcome the problem include process improvement and the use of modified microorganisms that produce the required hemicellulose enzymes (Lu and Mosier, 2007; Haan et al., 2007).

Enzymes

Lignocellulosic enzymes, mainly from fungi and bacteria, are important commercial products of LCW bio-processing used in many industrial applications including chemicals, fuel, food, brewery and wine, animal feed, textile and laundry, pulp and paper and agriculture

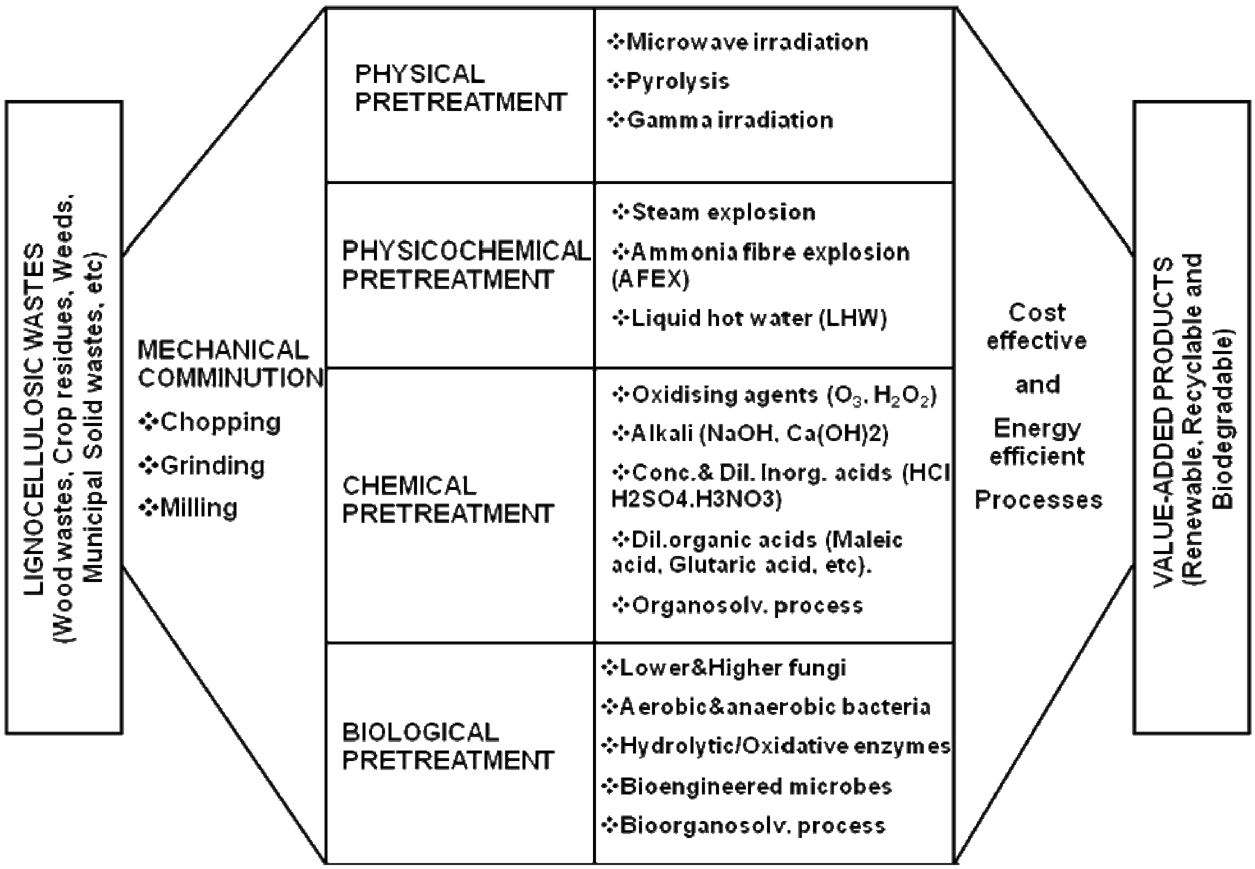

Figure 1. A summary of various methods used in the pretreatment of lignocellulosic wastes.

(Howard et al., 2003). Overall, extracellular enzymes are secondary metabolic products released in the presence of inducers at N-limited media (Mtui and Nakamura, 2007). They include hydrolytic enzymes such as cellulases; hemicellulases and pectinases; degradative enzymes like amylases, proteases; and ligninolytic enzymes like laccases, peroxidases and oxidases. Cellulases production from LCW has been extensively studied (Jecu, 2000; Emtiazi and Nahvi, 2000; El-hawary and Mostafa, 2001; Ögel et al., 2001; Raj and Singh, 2001; Ojumu et al., 2003; Wen et al., 2005; Muthuvelayudham and Viruthagiri, 2006; Pothiraj et al., 2006; Daroit et al., 2007; Gao et al., 2008). Phytases, mannanases and amylases are also produced by microorganisms using LCW as the main feedstock (Bhavsar et al., 2008; Mabrouk et al., 2008).

On the other hand, hemicellulolytic enzymes, mainly xylanases, are produced from a wide range of LCW biomass (Abdel-Sater and El-Said 2001; Rezende et al. 2002; Pandey and Pandey, 2002; Isil and Nilufer, 2005; Haq et al., 2006; Elisashvili et al., 2006; Dobrev et al., 2007; Mohana et al., 2008). Pectinases such as endo-polygalacturonase (endo-PG), exo-polygalacturonase (exo-PG) and pectin liase are mainly produced from solid state fermentation processes utilizing agricultural residues (Silva et al., 2005; Botella et al., 2005, 2007), while protease has been produced by *Penicillium janthinellum* in submerged cultures (Oliveira et al., 2006).

Among the ligninases produced from LCW, laccases are the mostly studied (Nazareth and Sampy, 2003; Moldes et al., 2003, 2004; Couto et al., 2006; Couto and Sanrománа, 2006; Mishra and Kumar, 2007; Alcántara et al., 2007; Minussi et al., 2007), followed by Manganese peroxidase and lignin peroxidase (Couto et al., 2001, 2003; Wuyep et al., 2003; Velázquez-Cedeño et al., 2004; Couto and Sanrománа, 2005; Alam et al., 2005; Asgher et al., 2006; Songulashvili et al., 2007; Elisashvili et al., 2008).

Very high enzyme activities (31,786 U/L) have been reported when the experiments are carried out under optimal conditions (pH 5.5 - 6: temperature 30 - 45°C) (Rosales et al., 2007). Recovery of pure enzymes is achieved through 50 - 80% $(NH_4)_2SO_4$ saturation followed by chromatographical purification techniques (A-el-Gammal et al., 2001; Mtui and Nakamura, 2008). Several efforts have been made to increase the production of enzymes through strain improvement by mutagenesis and recombinant DNA technology. Cloning and sequencing of the various genes of interest could economize the enzymes production processes (Kumar et al., 2008).

Biofuels

Worldwide, there is a growing concern over the fossil oil prices increase, the security of the oil supply and the negative impact of fossil fuels on the environment, particularly greenhouse gas emissions (Hahn-Hägerdal et al., 2006). Conversion of LCW to biofuels provides the best economically feasible and conflict-free second-generation renewable alternatives (Rubin, 2008). Significant advances have been made towards bioconversion of plant biomass wastes into bioethanol, biodiesel, biohydrogen, biogas (methane).

Production of ethanol from sugars or starch from sugarcane and cereals, respectively, impacts negatively on the economics of the process, thus making ethanol more expensive compared with fossil fuels. Hence, the technology development focus for the production of ethanol has shifted towards the utilization of residual lignocellulosic materials to lower production costs (Howard et al., 2003). Currently, research and development of saccharification and fermentation technologies that convert LCW to reducing sugars and ethanol, respectively, in eco-friendly and profitable manner have picked tempo with breakthrough results being reported (Lin and Tanaka, 2006; Prasad et al., 2007; Patel et al., 2007; Pasha et al., 2007; Tahezaden and Karimi, 2007; Sánchez and Cardona, 2008). Ethanol yield of 6 - 21% has been obtained through fermentation of agricultural and municipal residues (Akin-osanaiye et al., 2005; Mtui and Nakamura, 2005; Sjöde et al., 2007; Li et al., 2007; Cara et al., 2008; Sørensen et al., 2008). While microaeration enhances productivity of bioethanol from LCW using ethanologenic *E. coli* (Okuda et al., 2007), simultaneous saccharification and fermentation (SSF) using recombinant *Saccharomyces cereviasiae* result to as high as 62% of the theoretical value (Itoha et al., 2003). The principal benefits of performing the enzymatic hydrolysis together with the fermentation, instead of in a separate step after the hydrolysis, are the co-fermentation of both hexoses and pentoses during SSF, reduced end-product inhibition of the enzymatic hydrolysis and the reduced investment costs (Kádár and Réczey, 2004; Olofsson et al., 2008). Life cycle assessment (LCA) shows that bio-ethanol from LCW results to reductions in resource use and global warming (von Blottnitz and Curran, 2007). The long-term benefits of using waste residues as lignocellulosic feedstocks will be to introduce a sustainable solid waste management strategy for a number of lignocellulosic waste materials; contribute to the mitigation in greenhouse gases through sustained carbon and nutrient recycling; reduce the potential for water, air, and soil contamination associated with the land application of organic waste materials; and to broaden the feedstock source of raw materials for the bio-ethanol production industry (Champagne, 2007).

Biodiesel is a renewable fuel conventionally prepared by transesterification of pre-extracted vegetable oils and animal fats of all resources with methanol, catalyzed by strong acids or bases (Liu and Zhao, 2007). They are fatty acid methyl or ethyl esters used as fuel in diesel engines and heating systems (Ito et al., 2005). Production of biodiesel from lignocellulosic residues such as olive oil wastes has been a subject of research towards improving the thermal waste treatment systems and cleaner energy production (Arvanitoyannis et al., 2007a, 2007b). Since the current supplies from LCW based oil crops and animal fats account for only approximately 0.3%, biodiesel from algae is widely regarded as one of the most efficient ways of generating biofuels and also appears to represent the only current renewable source of oil that could meet the global demand for transport fuels (Schenk et al., 2008).

Hydrogen has been considered a potential fuel for the future since it is carbon-free and oxidized to water as a combustion product (Najafpour et al., 2004). While conventional burning or composting seem to be the most cost-effective hydrogen production methods, bacteria such as *Enterobacter aerogenes* and *Clostridium* sp isolates can convert saccharified LCW biomass into biohydrogen (Ito et al., 2005). Biohydrogen production from agricultural residues such as olive husk pyrolysis (Ça lar and Demirba, 2002); conversion of wheat straw wastes into biohydrogen gas by cow dung compost (Fan et al., 2006); bagasse fermentation for hydrogen production (Singh et al., 2007) generate up to 70.6% gas yields. System optimization for accessibility of polysaccharides in LCW and the use of genetically efficient bacterial strains for agrowaste-based hydrogen production seem to be the ideal option for clean energy generation. Hydrogen gereration from inexpensive abundant renewable biomass can produce cheaper hydrogen and achieve zero net greenhouse emissions (Zhang et al., 2007).

Biogas production from lignocellulosic materials is a steady anaerobic process where methane rich biogas comes mostly from hemicellulose and cellulose. Anaerobic biomethane production is an effective process for conversion of a broad variety of agricultural residues to methane to substitute natural gas and medium calorific value gases (Demirbas and Ozturk, 2005). Biogas containing 55 - 65% methane can be produced from jute caddis - a lignocellulosic waste of jute mills by anaerobic fermentation, using cattle dung as sole source of inoculum (Banik, 2004). Anaerobic digestion of poultry droppings, cow dung and corn stalk can give up to 137.16 L of biogas from 0.28 m^3 digester (Anozie et al., 2005). Mesophilic aerobic pretreatment to delignify sisal pulp waste prior to its anaerobic digestion has been shown to improve methane yields (Mshandete et al., 2005, 2008).

Overall, the success of biofuels production from LCW is dependent on the optimal performance and cost effectiveness of pretreatment and product generation processes.

Organic acids

Organic acids are some of the products of ligninolytic residues fermentations via environmentally friendly integrated processes. Volatile fatty acids including acetic acid, propionic acids and butyric acid are produced from a wide range of LSW such as cereal hulls (Jin et al., 2002, 2004, 2006); bagasse residues (Henrique et al., 2005); food wastes (Lim et al., 2008) and sisal leaf decortications residues (Mshandete et al., 2008). In addition, lactic acid is produced from waste sisal stems (Muruke et al., 2006), sugarcane bagasse (Adsul et al., 2007) and kitchen waste (Ohkouchi and Inoue, 2007) by using *Lactobacillus* isolates. Furthermore, formic acid, levulinic acid, citric acid, valeric acid, caproic acid and vanillinic acid are obtainable from bioprocessing of LCW (Olson, 2001; Chaudhary and Sharma, 2005; Mshandete et al., 2008; Ibrahim et al., 2008). Overall, organic acids production requires batch or continuous incubation conditions, the average reaction parameters being 35°C, pH 6.0, hydraulic retention time (HRT) of up to 8 days and organic loading rates of 9 g/l d. Product yields of up to 39.5 g/l have been reported (Lim et al., 2008).

Compost

Compost, a nutrient-rich, organic fertilizer and soil conditioner, is a product of humification of organic matter. This process is aided by a combination of living organisms including bacteria, fungi and worms which transform and enhance lignocellulosic waste into humic-like substances (Eyheraguibel et al., 2008). Vermicomposting is the bio-oxidation and stabilization of organic matter involving the joint action of earthworms and microorganisms, thereby turning wastes into a valuable soil amendment called vermicompost (Benitez et al., 2005; Aira et al., 2006, 2007). Substrates suitable for making humus–rich compost include cereal straw and bran (Hart et al., 2003); urban wastes (Taiwo and Oso, 2004); water hyacinth (Chatterjee et al., 2005); lemon tree prunings, cotton waste and brewery waste (García-Gómez et al., 2005); horticultural wastes (Wen-Jing et al., 2004, Lopez et al., 2006); olive, palm and grape wastes (Salètes et al., 2004; Alburquerque et al., 2006; Cayuela et al., 2006; Arvanitoyannis et al., 2007a). While bacteria inoculants such as *Bacillus shackletonni*, *Streptomyces thermovulgaris* and *Ureibacillus thermosphaericus* are used to improve the composting process (Vargas-Garcı et al., 2007), ligno-cellulolytic fungi inocula (e.g. *Trichurus spiralis*) may also be used in a pretreatment process before composting in order to reduce the resistance of the substrate to biodegradation (Hart et al., 2003; Vargas-García et al., 2007). A new earthworm strain of *Perionyx sansibaricus* is able to humify a substrate combination of guar gum industrial waste, cow dung and saw dust (Suthar, 2007). Composting can, therefore, be considered as a low-cost technology to convert agro-industrial LCW into value-added biofertilizers.

Biocomposites

Biodegradable polymers constitute a loosely defined family of polymers that are designed to degrade through the action of living organisms. Such commercially available biodegradable polymers are polycaprolactone, poly (lactic acid), polyhydroxyalkanoates, poly (ethylene glycol), and aliphatic polyesters like poly (butylene succinate) (PBS) and poly (butylene succinate-co-butylene adipate) (Tserki et al., 2006). Lignocellulosic material-thermoplastic polymer composites are among the emerging products of LCW. In most cases, lignocellulosic biomass flour is used as the reinforcing filler and polypropylene as the thermoplastic matrix polymer to manufacture particle-reinforced composites (Yang et al., 2004). Natural fibres from LCW are considered to be of low-cost by-products, environmentally friendly and practically sustainable raw materials (Georgopoulos et al., 2005). Evaluations of LCW fiber plastic composites utilizing wood fibre wastes (Bhattacharyya and Jayaraman, 2003; Yuan et al., 2004; Schilling et al., 2004; Ashori, 2008); wheat and rice straw (Digabel et al., 2004; Yang et al., 2004a); jute/cotton, sisal/cotton and ramie/cotton hybrid fabrics (Mishra et al., 2004; Alsina et al., 2005; Jacob et al., 2006); non-wood plant fibres (Ndazi et al., 2006); waste newsprint paper (Madani et al., 2004; Baroulaki et al., 2006); flax and hemp (Tserki et al., 2006); oil palm wastes (Shaji et al., 2006; John et al., 2008); cotton gin waste (Bourne et al., 2007); banana fibres (Pothan et al., 2007); cereal husks (Yang et al., 2004b, 2007; López et al., 2007); tissue paper wastes and corn peels (Lertsutthiwong et al., 2008); bagasse (Habibi et al., 2008) and nanofibers from the agricultural residues (Alemdar and Sain, 2008) have shown that such composites are suitable for making products that have improved biodegradability, mechanical strength, thermal stability, electrical conductivity and recyclability.

Treated LCW wastes are also used in the construction industry for manufacturing of light-weight agro-gypsum panels (Basta et al., 2002) and lightweight sand concretes (Reis, 2006; Bederina et al., 2007) with improved structural and thermal properties. Biocomposites are very promising in producing sustainable current and future green materials to achieve durability without using toxic chemicals. The challenge facing the biocomposite industry is to make materials that have better rubber/fiber interface, improved wettability and compatibility.

Food and feed

Bioconversion of lignocellulosic agro-residues through mushroom cultivation and single cell protein (SCP) production offer the potential for converting these residues into protein-rich palatable food and reduction of the environmental impact of the wastes. Mushrooom cultivation provides an economically acceptable alternative for the production of food of superior taste and quality which does not need isolation and purification (Israilides and Philippoussis, 2003; Philippoussis et al., 2007). Cultivation of edible mushrooms such as *Lentinus* spp, *Lentinula* spp, *Leonotis* spp, *Pleurotus* spp, *Agaricus* spp, *Agrocybe* spp*, Volvariella* spp, *Lentinus* spp and *Grifola* spp is achievable on a wide range of LCW substrates such as wood waste, corncob meal, wheat straw, barley straw, soybean straw, cereal bran, cotton waste, sorghum stalk, banana pseudostem, hazelnut husks, waste tea leaves, dry weed plants, peanut shells, waste paper and olive mill wastewater (Morais et al., 2000; Philippoussis et al., 2001; Yildiz et al., 2002; Oku, 2004; Kalm and Sargın, 2004; Silva et al., 2005; Özçelika and Pekşen, 2007; Peker et al., 2007; Das and Mukherjee, 2007; Akyüz and Yildiz, 2008; Gaitán-Hernández and Salmones, 2008; Rani et al., 2008). Mushrooms with increased number of fruit bodies and high contents of protein and total carbohydrates are obtained when LCW substrates are used in combination.

On the other hand, SCP production from LCW offers a potential substrate for conversion of low-quality biomass into an improved animal feed and human food. SCP is the protein extracted from cultivated microbial biomass. It can be used for protein supplementation of a staple diet by replacing costly conventional sources like soymeal and fishmeal to alleviate the problem of protein scarcity. Moreover, bioconversion of agricultural and industrial wastes to protein-rich food and fodder stocks has an additional benefit of making the final product cheaper (Anupama and Ravindra, 2000). Removal of nucleic acids and toxins from SCP is key to ensure the safety of food and feed. Among the SCP obtained from LCW using agricultural wastes as the main growth media, *Saccharomyces cerevisiae*, *Trichoderma reesei* and *Kluyveromyces marxianus* top the list (Robinson and Nigam, 2003; Chaudhary and Sharma, 2005). SCP yield of 51 and 39.4% efficiency of conversion of beet-pulp into protein has been reported ffrom the above strains. Solid state fermentation of LCW seems to be the most preferred culturing method, while cloning is being considered as a suitable technique for improvement of SCP production (Anupama and Ravindra, 2000).

Medicines

LCW provides a suitable growth environment for mushrooms that comprise a vast source of powerful new pharmaceutical products. In particular, *Lentinula edodes*, *Tremella fuciformis* and *Ganoderma lucidum* contain bioactive compounds such as anti-tumor, anti-inflammatory, anti-virus and anti-bacterial polysaccharides. Moroever, they contain substances with immunomodulating properties, as well as active substances that lower choresterol (Israilides and Philippoussis, 2003; Philippoussis et al., 2007; Zhang et al. 2007). Future prospects for research on bioactive compounds from fungi grown on such cheap and ubiquitous substrates look bright and could lead to breakthroughs in the search for antibacterial, antiviral and anticancer chemotherapies.

Biosorbents

Adsorbents obtained from plant wastes are feasible replacements for costly conventional methods of removing pollutants such as heavy metals ions, dyes, ammonia and nitrates from the environment. The use of lignocellulosic agrowastes is a very useful approach because of their high adsorption properties, which results from their ion-exchange capabilities. Agricultural wastes can be made into good sorbents for the removal of many metals, which would add to their value, help reduce the cost of waste disposal, and provide a potentially cheap alternative to existing commercial carbons (Krishnani and Ayyappan, 2006). Chemically modified plant wastes such as rice husks/rice hulls, spent grain, sugarcane bagasse/fly ash, sawdust, wheat bran, corncobs, wheat and soybean straws, corn stalks, weeds, fruit/vegetable wastes, cassava waste fibres, tree barks, azolla (water fern), alfalfa biomass, coirpith carbon, cotton seed hulls, citrus waste and soybean hulls show good adsorption capacities for Cd, Cu, Pb, Zn and Ni (Ahmedna et al., 2004; Basso et al., 2004; Dupont et al., 2005; Harman et al., 2007; Šćiban et al., 2008; Ngah et al., 2008; Zubair et al., 2008). They are usually modified with formaldehyde in acidic medium, NaOH, KOH/K_2CO_3 and CO_2, or acid solution or just washed with warm water (Tsai et al., 2001; Šcibanet al., 2008). Scanning electron micrographs with energy spectra shows that heavy metals are immobilized via two possible routes: adsorption and cation exchange on hypha, and the chelation by fungal metabolite (Huang et al., 2008).

LCW have also been shown to be able to adsorb dyes from aqueous solutions. Adsorption of reactive dyes by sawdust char and activated carbon (Gan et al., 2004); ethylene blue by waste *Rosa canina* sp. seeds (Gürses et al., 2006); anionic dyes by hexadecyltrimethylammonium-modified coir pith (Namasivayam and Sureshkumar, 2006); and methylene red by acid-hydrolysed beech sawdust (Batzias and Sidiras 2007) have been reported. Ammonia and nitrate removal by using agricultural waste materials as adsorbents or ion exchangers have also been studied (Orlando et al., 2002; Kishore et al., 2006). Prehydrolysis enhances the adsorption properties of the original LCW material due to the removal of the hemicelluloses during sulphuric acid treatment, resulting in the ‘opening’ of the lignocellulosic matrix’s structure, the increasing of the surface area and the activation of the material’s surface owing to an increase in the number of dye binding sites (Batzias and Sidiras, 2007). The main value-added products from LCW are generally summarized in Figure 2.

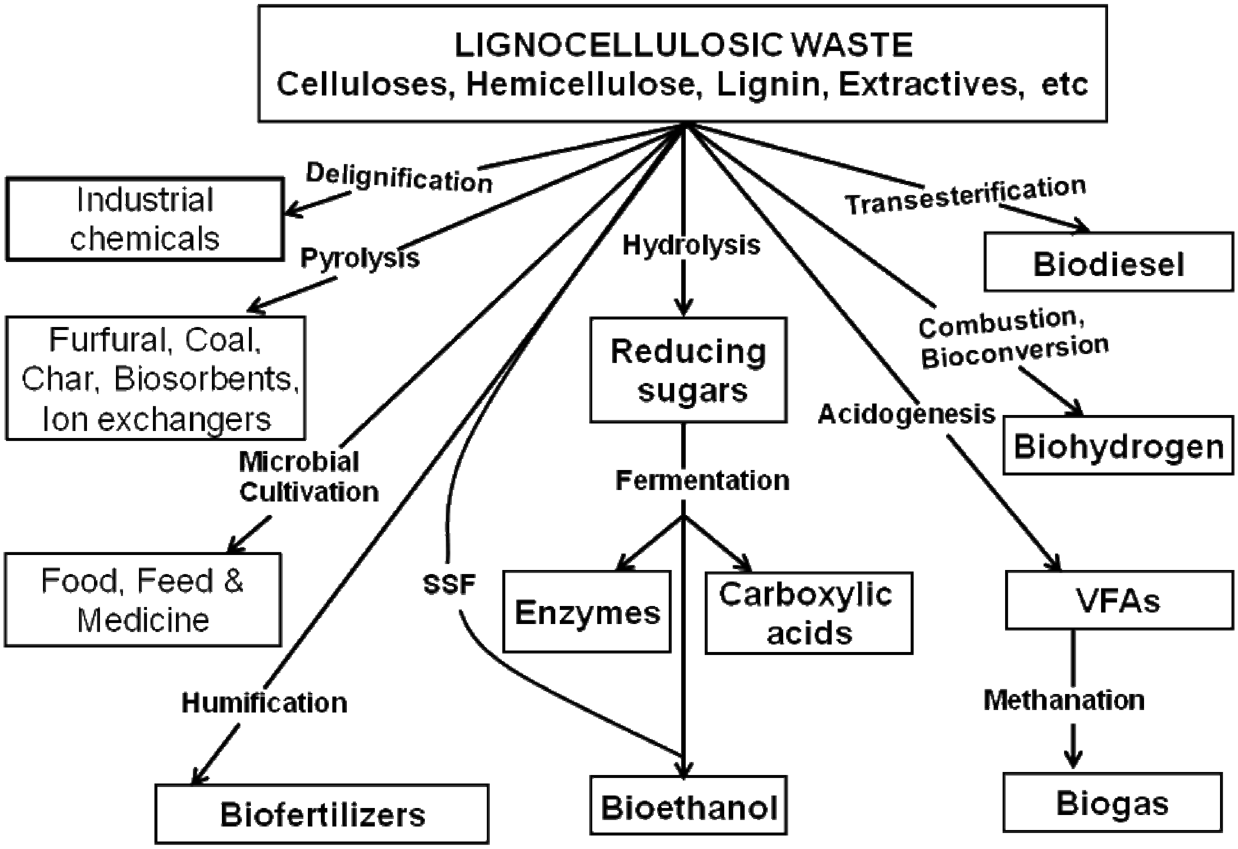

Figure 2. The main value-added products from lignocellulosic wastes (SSF=simultaneous fermentation and saccharification, VFAs = volatile fatty acids).

TECHNO-ECONOMIC EVALUATION

Technologies that are being developed for commercial pretreatment and value addition of LCW face technical and economical impediments. Therefore, cost effective technical innovations that allow cost-effective conversion of biomass into fuels and chemicals are mandatory. These technologies include low-cost thermochemical pretreat-ment, highly effective enzymes and efficient and robust fermentative microorganisms (Gray et al., 2006; Cardona and Sánchez, 2007; Vertès et al., 2008). The choice of appropriate feedstocks and processes ought to consider those which consume less electricity, produce fewer emissions in total, and has less of a human health impact (Kemppainen and Shonnard, 2005).

The high cost of enzymes presents a significant barrier to commercialization of bio-based products. In the simplest terms, the cost is a function of the large amount of enzyme protein required to break down polymeric sugars in cellulose and hemicellulose to fermentable monomers. In recent years, significant effort has been expended to reduce the cost by focusing on improving the efficiency of known enzymes, identification of new, more active enzymes, creating enzyme mixes optimized for selected pretreated substrates, and minimization of enzyme production costs (Merino and Cherry, 2007).

Ethanol's future role as a fuel hinges on several factors including feedstock availability, processing costs and supportive political framework. Improvements in pre-treatment and advances in biotechnology, especially through process combinations can bring the ethanol production overall process efficiency to 68%. Also, a combined effect of higher hydrolysis-fermentation efficiency, lower specific capital investments, increase of scale, cheaper biomass feedstock costs and using genetically engineered microorganisms that can convert xylose and/or pentose to ethanol can greatly improve ethanol production efficiency and reduce the cost of the production (Sun and Cheng, 2002; Carlo et al., 2005). Processes that produce only ethanol from lignocellulosics display poor economics. The large market for ethanol makes it possible to achieve economies of scale that reduce sugar costs, and co-producing chemicals promises greater profit margins or lower production costs for a given return on investment. For the large processing plants, the production costs are significantly reduced compared to small plants (Wyman, 2003; Murphy and McCarthy, 2005). Yield improvements in all major steps of LCW processing would enable lower capital re-quirements, thus improving the economics and lowering investment risk (Bohlmann, 2006). Bioproducts from LCW will continue to be the keystone of industrial biotechnology-based economy whereby biorefineries leverage common raw materials and unit operations to integrate diverse processes to produce demand-driven product portfolios (Otero et al., 2007). High product yields and less energy-demanding processes could be achieved by increasing the dry matter content resulting in higher products, and thereby improving the overall process economy (Sassner et al., 2008).

FUTURE TRENDS

Although pretreatment systems and the concomitant release of bio-products from LCW have been greatly improved by new technologies, there are still challenges that need further investigations. These challenges include development of more efficient pretreatment and production technologies, bioprospecting and development of stable genetically engineered microorganisms, improved gene cloning and sequencing technologies and enhancement of productions based on economies of scale for more efficient and cost effective conversions of LCW into value-added products.

So far, lignocellulosic biomass has been the most promising economically viable and renewable source of biohydrogen and biodiesel. However, the second generation microalgal systems seem to be more advantageous in that they: (1) have a higher photon conversion efficiency (as evidenced by increased biomass yields per hectare): (2) can be harvested batch-wise nearly all-year-round, providing a reliable and continuous supply of oil: (3) can utilize salt and waste water streams, thereby greatly reducing freshwater use: (4) can couple CO_2-neutral fuel production with CO_2 sequestration: (5) produce non-toxic and highly bio-degradable biofuels (Schenk et al., 2008). Therefore, extensive research is now being directed toward that end. Plant fibers as fillers and reinforcements for polymers are currently the fastest-growing type of polymer additives. Nanobiotechnology seems to take charge as far as the use of LCW nanofibres in plastic composites is concerned (Alemdar and Sain, 2008). It is envisaged that nano materials from renewable biowastes will be the main focus of future research.

Conclusion

This work has attempted to take a broad analysis of most of the research done worldwide in the past nine years regarding the enormous diversity of value-added products from pretreated lignocelluloses wastes (LCW). Various pretreatment and production systems providing technical and economic feasibility to harness the renewable materials while at the same time cleaning up the environment have been highlighted. Physicochemical and biological pretreatment systems appear to be the mostly preferred methods, while the use of treated LCW as bio-based adsorbents of pollutants, and the production of degradable biofuels and biopolymers from LCW have drawn a lot of research interest. Current and future research trends are directed towards the developments and applications of engineered organisms to tackle the challenges encountered from using conventional naturally occurring strains. At the same time, production of *nano* materials from pretreated LCW provides bright future prospects in the biocomposites industry. Although the main challenge facing LCW pretreatment and value-addition industry over the next few years will be to reduce processing and production costs, this work looked at studies that suggest the ways out of this problem. The review serves as a valuable reference material for a wide range of scientists and technologists in the relevant fields.

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