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REVIEW ARTICLE

Challenges in developing strategies for the valorization of lignin—a major pollutant of the paper mill industry

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Abstract

Apart from protecting the environment from undesired waste impacts, wastewater treatment is a crucial platform for recovery. The exploitation of suitable technology to transform the wastes from pulp and paper industries (PPI) to value-added products is vital from an environmental and socio-economic point of view that will impact everyday life. As the volume and complexity of wastewater increase in a rapidly urbanizing world, the challenge of maintaining efficient wastewater treatment in a cost-effective and environmentally friendly manner must be met. In addition to producing treated water, the wastewater treatment plant (WWTP) has a large amount of paper mill sludge (PMS) daily. Sludge management and disposal are significant problems associated with wastewater treatment plants. Applying the biorefinery concept is necessary for PPI from an environmental point of view and because of the piles of valuables contained therein in the form of waste. This will provide a renewable source for producing valuables and bio-energy and aid in making the overall process more economical and environmentally sustainable. Therefore, it is compulsory to continue inquiry on different applications of wastes, with proper justification of the environmental and economic factors. This review discusses current trends and challenges in wastewater management and the bio-valorization of paper mills. Lignin has been highlighted as a critical component for generating valuables, and its recovery prospects from solid and liquid PPI waste have been suggested.

Keywords Biorefinery approach · Carbon dots · Lignin nanoparticles · Pulp and paper industry wastewater · Paper mill sludge (PMS) · Microbial treatment

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Highlights
- Biorefinery approach for valorization of paper mill wastes.
- Conventional treatment strategies.
- Need for microbial treatment of paper mill wastewater.
- Biological methods of solid waste utilization.
- Usage of lignin into specialized valuables.

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Introduction

Manufacturing enterprises have a significant role in strengthening the financial system of any nation. During the manufacturing process of different products, these industries yield harmful waste in the form of gasses, solids, and liquids which deteriorate the quality of the environment and directly impact human health (Murillo-Luna et al. 2011). As industrial development began in the late twentieth century, industrial wastewater generation increased significantly. Industrial wastewater contains many new organic compounds generated yearly due to industrial activities. Technological changes in the manufacturing unit also change the combination of discharged and, in turn, wastewater characteristics (Tchobanoglous et al. 1991). Several compounds generated from industrial processes are strenuous and expensive to treat by traditional wastewater processes.

Interestingly, the wastewater also represents a repository of valuable by-products. However, the trend toward converting waste into valuable materials is not practiced appreciably in developing countries (Bajwa et al. 2019). Looking at the global scenario of environmental consciousness, it has become essential for all industries to use each component holistically and follow a biorefinery approach. The conventional use of physical and chemical methods can lead to rising energy utilization and the generation of secondary pollutants, for which biological methods are coming up as a choice.

Among the various industrial sectors, the pulp and paper industry has gained the pace of development. Several governments of leading nations have recognized it as a high-priority industrial sector (Kamali and Khodaparast 2015). It is known that this industry, when processing lignocellulosic biomass and chemicals, requires a large volume of water at each processing stage and generates enormous amounts of pulp mill effluent (liquid waste) and sludge (solid waste) (Patel et al. 2017). Therefore, the disposal of both parts are the two main environmental issues. Most paper pulp mill effluent is characterized by alkaline pH, dark color, high chemical oxygen demand (COD), and suspended solids that result in acute and chronic toxicity to aquatic biota (Patel et al. 2017; Majumdar et al. 2019b, a).

The complex and recalcitrant lignin with chlorinated lignin derivatives present in the paper pulp mill effluent is the major pollutant and causative agent for genetic mutations in exposed organisms (Lindholm-Lehto et al. 2015). Lignin imparts dark brown color, which severely impedes the natural process of photosynthesis by obstructing sunlight's penetration into the water. Various treatment methods based on physical (adsorption, microfiltration, and photoionization) and chemical (sedimentation, coagulation, oxidation, and ozonation) principles are used (Kamali and Khodaparast 2015). However, these methods have serious drawbacks, including generating secondary pollution with costly mitigation processes. It seems that if this wastewater is used for the extraction of lignin, it cannot only help in the reduction but also in the production of valuable lignin derivatives. Now-a-days lignin can be converted into lignin nanoparticles and even carbon dots (H. Liu et al. 2018a, b; T. Liu et al. 2020). Recently these compounds have been found to indicate several biomedical and industrial applications, such as drug delivery systems, imaging dye, and many more (Cheng et al. 2020; Rai et al. 2017).

This review appraises the possibilities of producing valuables from both solid and liquid waste of the pulp and paper industry for various purposes with the overall utilization of the waste materials using biological routes. This review addresses the strategies developed firstly for the effluent valorization and second for the use of paper mill sludge (PMS), the solid waste of the same industry as a storehouse of essential commodities, majorly through the utilization of lignin.

Paper and pulp industry: the need of developing countries/threat and benefit

Paper plays a vital role in the evolution of our civilization, and it is tough to imagine modern life without paper. Although the SARS-CoV-2 pandemic has severely affected the world, the global consumption of paper and cardboard in 2020 is still 399 million metric tons and is expected to increase to 461 million metric tons by the end of the next decade (Tiseo 2021). India ranks 20th in world paper production, with China, the USA, and Japan leading worldwide (Malaviya and Rathore 2007b). The pulp and paper industries are among the most environmentally concerned, especially in countries like India (Bajpai 2015a, b). The annual paper production in India is around 10.11 million tons, estimated to be 2.6% of the world's overall production (Working group report 2011).

Furthermore, India possesses one of the fastest-growing pulp and paper markets, with an annual growth rate of > 10% per capita consumption (Bajpai 2015a, b). In India, 60–70% of raw materials used for papermaking are hardwood, bamboo fibers, and agro-wastes, whereas 30–40% are from recycled sources. Generally, the demand for paper and board is growing annually by 5–6% globally (Bhatnagar 2015). The economic profits of the pulp and paper industry have made it a necessary industrial sector globally. However, the industry is among the five major pollution contributors to precious water bodies (Pokhrel and Viraraghavan 2004). It releases millions of tons of contaminated effluent into the aquatic resources yearly (Rigol et al. 2003). Every year approximately 100 million kg of harmful contaminants are generated from the paper and pulp industry (Gopal et al. 2019).

As per our country's scenario, India's paper manufacturing units produce 11 million tons per annum and generate 40–50 kg of sludge (dry) per tonne of paper (Working Group report of the 12th Fifth Year Plan). The abundant amount of paper mill sludge (PMS) is mainly considered waste and usually disposed of or burnt off, emitting greenhouse gasses (Fang et al. 2017). Disposing of or using sludge has long been challenging for the pulp and paper industry (Geng et al. 2007). The impetus toward resource utilization and reduced landfills has emphasized the research into the constructive use of PMS (Deeba et al. 2016). The abundantly available PMS has been explored for incorporation into building materials, cement, and ceramics (Coutts 2005; Frías et al. 2015) attributed to its compositional feature of higher cellulose content imparting mechanical strength. The lignocellulosic composition of PMS reminds us of the generation of valuables from the same via utilization of components that will be again a proper biorefinery strategy for the pulp and paper industry.

The primary processes in the paper and pulp industries are debarking, pulping, separating the pulp from cooking liquor, bleaching, stock preparation, and producing final paper products (Bajpai 2015a, b). The primary and foremost aim lies in the effective removal of the lignin seal to expose the cellulose structure for the conversion and formation of valuable products. Now-a-days there is a wide range of conventional physicochemical technologies used for the delignification of lignocellulose biomass. A modern paper mill can operate using less than 6 m^3 fresh water per ton of paper. In contrast, older mills may require more water, even 60 m^3 per ton of paper (Mänttäri et al. 2004). More than 42% of the wastewater produced in the world is generated by the pulp and paper industry (Pokhrel and Viraraghavan 2004). It poses a significant threat to the environment and ignites the necessity of wastewater management. Moreover, the enormous amount of sludge generated as a result of the water treatment process is a significant source of soil pollution, leading to further alarming situations (Deeba et al. 2016). Except for a small percentage of the waste paper used in paper recycling, the rest is dumped or incinerated without any economic importance. In addition, till now not a single satisfactory waste recycling system is implemented, although this leading industrial sector enormously contributes to the country's economy. A biorefinery approach for this industry is the need of the hour. Wastewater and PMS, if utilized for the generation of valuables, can further contribute to the economy and reduce environmental hazards to a great extent.

Paper and pulp mill wastes: characteristics and potential hazards

Pulp and paper mill wastes are broadly classified into liquid and solid wastes each having its uniqueness. A dark-colored alkaline waste stream produced by paper mills, referred to as "black liquor," contains inorganic compounds and wood waste (Chandra et al. 2011). Kraft lignin and phenol derivatives are the primary environmental pollutants in black liquor produced by pulp and paper mills (Raj et al. 2014; Hooda et al. 2015). It is characterized by color, very high biological oxygen demand (BOD), and chemical oxygen demand (COD) due to lignin and its derivatives (Tiku et al. 2010). The pH ranges from 6.1 to 8.3, with the low pH being due to the result of the metabolism of fungal species and the bio-acid metabolism of the native microflora (Kesalkar et al. 2012). Total dissolved solids concentration in the wastewater generally ranges from 395 to 2500 mg/l, and COD varies from 480 to 4450 mg/l (Kumar et al. 2015a, b). In addition to the natural polymers (tannins, lignin), it also contains xenobiotic components produced or used during manufacturing process (chlorinated lignins, chlorinated resin acids, chlorinated phenols, chlorinated polyaromatic hydrocarbons compounds) (Chandra and Singh 2012) and high sodium concentration (Kesalkar et al. 2012). In addition to that, recycled wastewater contains 2,4,7,9-tetramethyl-S-decyne-4,7-diol surfactant used in paints and printing inks (Kamali and Khodaparast 2015) and non-biodegradable organic substances including metal, sand, glass, and plastic components (Buyukkamaci and Koken 2010).

This wastewater, mainly discharged into the environment without adequate treatment, poses severe threats to aquatic, animal, and plant life. Large-size paper mills are generally located near perennial aquatic bodies, and the extracted alkali-soluble lignin fragments and their derivatives are discharged during the process (Chhonkar et al. 2000). Black liquor disposal generates a high concentration of persistent organic pollutants (POPs) as effluent, which harms aquatic flora and fauna (Kortekaas et al. 1998; Nie et al. 2013). According to numerous studies (Owens et al. 1994; Vass et al. 1996; Schnell et al. 2000; Lindström-Seppä et al. 1998; Leppänen and Oikari 1999; Johnsen et al. 1998; Ericson and Larsson 2000), adverse effects can include respiratory stress, mixed oxygenase activity, toxicity and mutagenicity, liver damage, or genotoxicity. They can also cause health hazards such as diarrhea, vomiting, headaches, nausea, and eye irritation in children and employees (Mandal 1996). Lignin and its derivatives released during delignification impart a dark brown color that makes it difficult for sunlight to penetrate the water (Karrasch et al. 2006). It is responsible for extreme adverse effects on aquatic flora and fauna and ultimately worsens human health through biomagnification in the food chain.

In addition, the solid waste generated from paper mill sludge (PMS) has potential threats. It is produced during deinking, pulping processes, and wastewater treatment. As studied, the extent and consistency of the solid waste rely on the raw resources used, the application of techniques, grade, and quality of the paper to be accomplished. Solid waste is generally moist and contains significant ash (Monte et al. 2009). Various gasses such as nitrogen oxides, hydrogen sulfides, sodium sulfides, methyl mercaptan, chlorine dioxides, and sulfur compounds are released from solid waste. These gasses and solid waste are responsible for chronic disorders, such as headaches and nausea. A trace of metals is also released into the water bodies as effluents. These have many unenthusiastic environmental and communal impacts and cause climate alteration (Sarma 2014). Sludge generation is mainly after primary and secondary wastewater treatment. Apart from these many other pollutants, such as grit bark from sorting and screening raw wood material, ash from coal-fired boilers and power generation systems, and lime sludge from a chemical recovery section of mills are produced (Sumathi and Hung 2006).

Conventional treatment strategies for paper pulp industry wastes

Physico-chemical treatment of papermill effluent

The effluent must be treated internally to reduce COD, BOD, and color. Treatment includes physical, chemical, physico-chemical, and biological processes (Tchobanoglous et al. 1991). Different physical, chemical, physicochemical, and electrochemical measures have been utilized to mitigate various industries' effluents. Suspended solids are removed in physical treatment, and wastewater is homogenized to regulate fluctuating flows (Buyukkamaci and Koken 2010). The wastewater is then chemically treated using coagulation, precipitation, adsorption, and coagulation techniques. Adsorbents such as chitosan, rice hull ash containing silica, rice hull ash without silica, ferric sulfate, and poly aluminum chloride have been used to remove pollutants from wastewater by several researchers (Buyukkamaci and Koken 2010; Kamali and Khodaparast 2015; Kumar et al. 2016). Advanced oxidation processes (AOPs) and membrane technologies have gained significant importance among the various methods, thus being highlighted in this review.

Advanced oxidation processes

AOP plays a significant role in paper and pulp waste treatment, specifically, using ozone and Fenton's reaction. Advanced oxidation processes (AOPs) are widely employed chemical treatments to deal with refractory organic pollutants (Babuponnusami and Muthukumar 2012b). The oxidation of complex compounds is carried out by non-selective hydroxyl radicals produced from Fenton's reagent, generated through a cascade of complex reactions (Babuponnusami and Muthukumar 2012a, b; Hussain et al. 2013). Fenton reagent's application had been proven to reduce COD value by 95% in industrial effluents containing phenolics, cyanides at optimum temperature, pH, $FeSO_4$ concentration, and H_2O_2 concentration (Amat et al. 2005; Sevimli 2005). Mandal et al. (2010) showed that when used in combination with *Thiobacillus ferrooxidans*, H_2O_2 concentration can be minimized and thus the cost. It is noticed that though effective, AOPs are quite cost-intensive processes, leading to the generation of undesirable by-products (Chaparro and Rueda-Bayona 2020; Liu et al. 2020).

Similarly, ozone oxidation can help in the oxidation of compounds in two ways: directly by reacting with dissolved substances or indirectly by hydroxyl radicals generated during the decomposition process (Amat et al. 2005). Ozonation proved effective in oxidizing chemicals in paper and pulp industry effluents such as catechol, vanillin, phenol, syringaldehyde, trichlorophenol, chlorophenol, eugenol, guaiacol, and derivatives of cinnamic acid. When assisted with UV rays at pH 6, this ozone can be removed by approximately 40% COD (Amat et al. 2005; Sevimli 2005). Though effective, the significant drawback is because of its short life, and the requirement of continuous ozonation makes this process costly.

Membrane technologies

Membrane technology has emerged as the preferred option to recover water from various wastewater streams for reuse (Asano and Cotruvo 2004). It has been proven that the membrane filtration technique has been found beneficial in treating the paper and mill industry. Microfiltration, followed by ultrafiltration and reverse osmosis, can provide high water recovery rates and excellent chemical composition (Pizzichini et al. 2005). Zhang (2012a) has shown that using a composite flocculant before reverse osmosis can reduce COD by up to 75%. Progress in wastewater treatment processes is obligatory for converting treated wastewater to re-usable for industrial, agricultural, and domestic purposes. However, such systems suffer serious drawbacks with technical and economic constraints mainly related to the retentate disposal and the membrane susceptibility to fouling (Obotey Ezugbe and Rathilal 2020).

Treatment of PMS: the solid waste

Several strategies have been taken to explore technologies that can help increase the digestibility of PMS in industries for its management. Among these, the various pretreatment technologies prior to anaerobic digestion have become more prominent (Meyer and Edwards 2014). These are broadly divided into physical, chemical, and biological methods (Fig. 1), as discussed below.

One of the most common modes of PMS pretreatment in industries is physical pretreatment, which consists of mechanical and thermal pretreatment. The main objective behind mechanical pretreatment is to disrupt cellulosic fraction crystallinity and reduce the degree of polymerization, making PMS more susceptible to enzymatic hydrolysis (Veluchamy and Kalamdhad 2017a, b, c, d). However, mechanical pretreatment is energy-consuming, high cost, and time-consuming. This strategy is lesser effective because it does not remove the lignin content from PMS biomass (Zheng et al. 2014). In thermal pretreatment, holocellulose hydrolysis can be achieved by different modes of heating over some time with rapid decompression (Veluchamy and Kalamdhad 2017a, b, c, d). Thermal pretreatment is highly effective in increasing the accessible surface area of cellulosic moiety and enhancing the susceptibility of cellulose to microbes and enzymes. However, the optimum parameters and magnitude of enhancement have been found to vary considerably (Wood et al. 2009). Among the other pretreatment methods, chemical pretreatment has gained significant attention. Paper industries use this pretreatment for PMS to delignify it to manufacture cellulose-based commodities or bioethanol and biogas. Various chemical methods are in use (Fig. 1), like acid and alkaline (thermal) hydrolysis, ozonation, and advanced oxidation methods (Veluchamy and Kalamdhad 2017a, b, c, d). These methods are applied for the pretreatment of PMS in paper industries for the removal of lignin and exposure of cellulosic fraction for the generation of valuables. Figure 2 represents the generation and various management methods of PMS in industries. However, environmental hazards generated through these processes remain the foremost hindrance in applying chemical pretreatment methods.

Need for biological treatments for paper industry wastewater and sludge

The physico-chemical techniques have severe hindrances, such as massive generation of sludge, huge capital investment, formation of secondary by-products, and dependence on energy sources limits its implementation (Banat et al. 1996). Due to the zero discharged policy, the industries involve innovative eco-friendly, sustainable, and efficient treatment practices. Nowadays, microbial technology is a much-admired option to treat wastewater (Chen et al. 2008). As studied, it is considered effective in combating lignin and other xenobiotic components found in industrial effluents (Rai et al. 2005). Various fungal species including basidiomycetes and ascomycetes along with bacterial strains are considered nature's original recyclers that could convert toxic organic compounds into harmless products. Exploration of the microbial diversity, particularly from the contaminated areas, offers a valuable platform for searching for organisms that can degrade a broad spectrum of pollutants in paper effluent (Kumar et al. 2017; Majumdar et al. 2019a, b; Beheraa et al. 2019). Biological processes have been noticed to be beneficial with respect to generation of reduced harmful by products and sludge. It is interesting that these processes not only require minimum fresh water and energy but it can also produce useful products such as pigments and enzymes, phenolics byproducts (Banat et al. 1996; Rai et al. 2005). The effective function of microbial process has been found to be associated with their inductive metabolic activity. Some bacterial and fungal species can convert hazardous pollutants into non-toxic end products (Priyadaarshinee et al. 2016; Majumdar et al. 2020a, b, c). In the bioremediation process, the potentiality of these microbes to decolorize, degrade, and detoxify lignin and other xenobiotics is harnessed. Microorganism-based remediation approaches have meticulously been scrutinized as potential tools to remove and degrade diverse kinds of hazardous environmental contaminants.

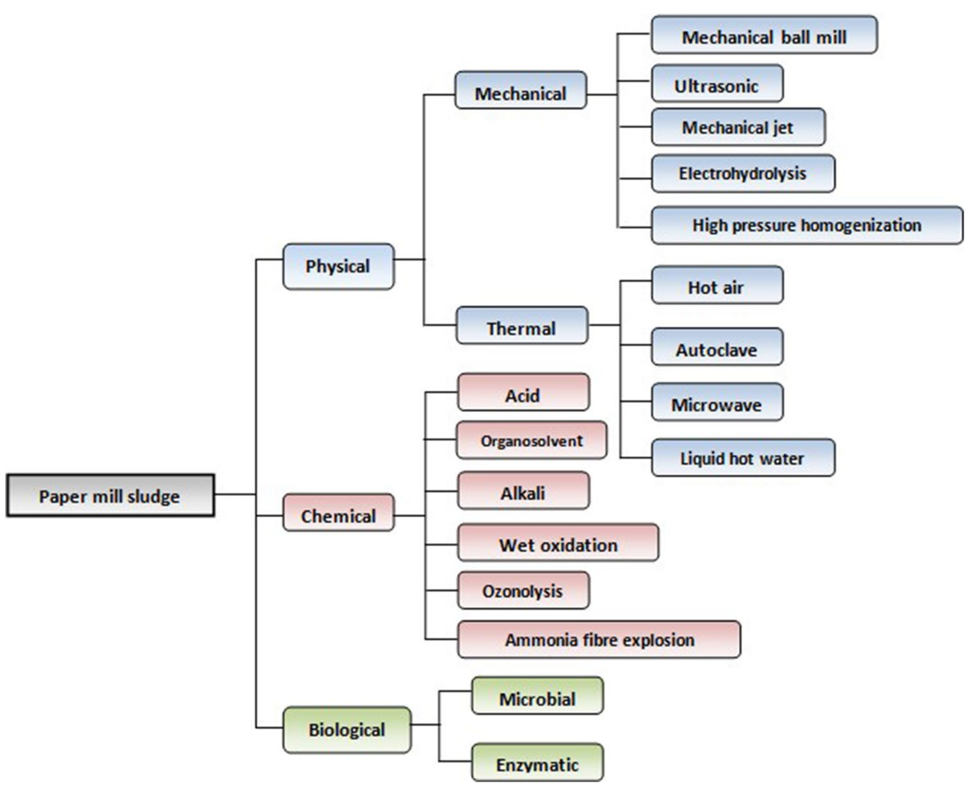

Fig. 1 Various pretreatment methods of PMS (physical, chemical, and biological) in pulp and paper industries (Veluchamy and Kalamdhad 2017b, c, d, a)

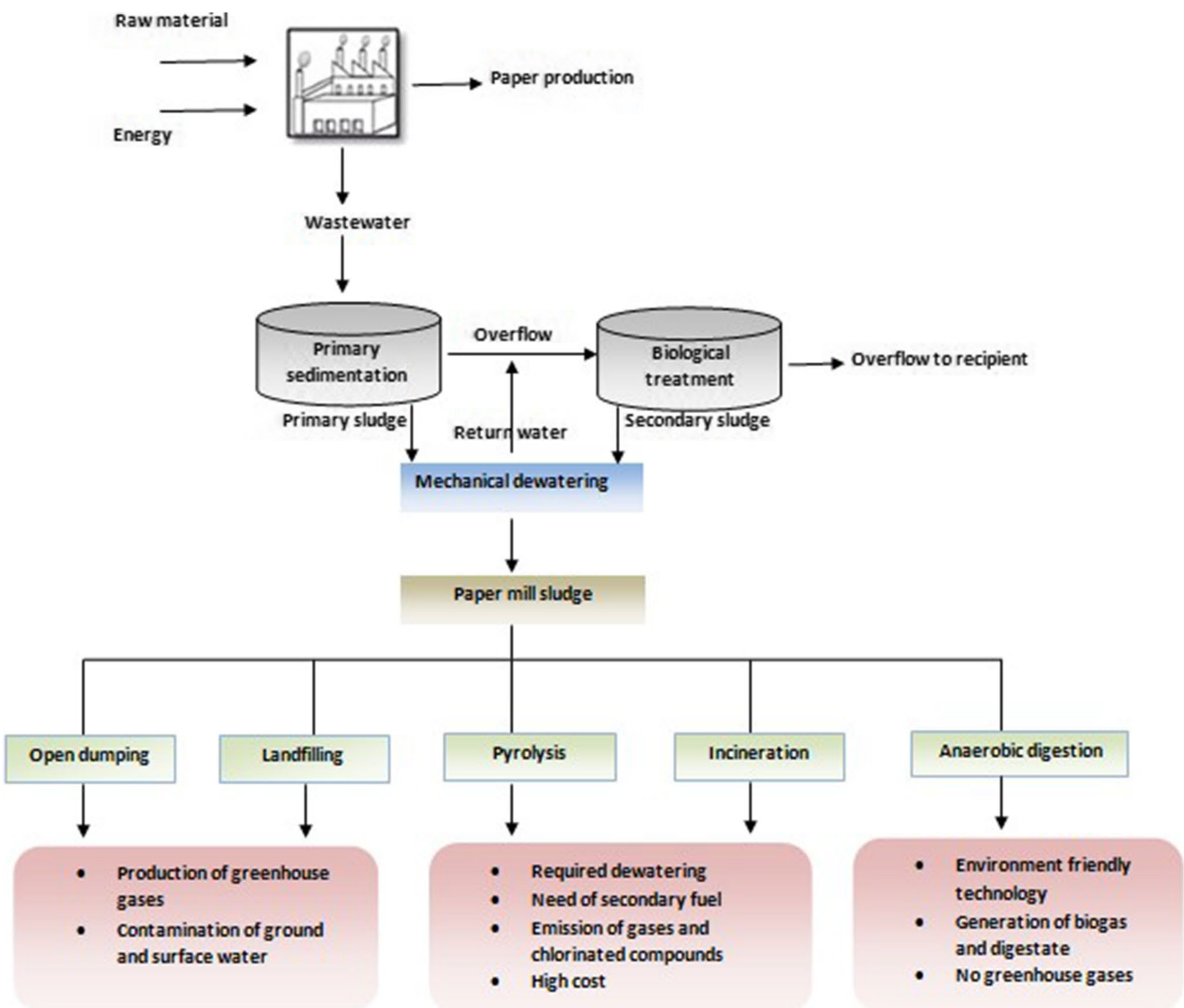

Fig. 2 Generation and management strategies of paper mill sludge in pulp and paper industries (Veluchamy and Kalamdhad 2017b, c, d, a)

Table 1 represents the various microbial species and their lignin degradation potentials.

Role of fungal species

In literature, many fungi have been widely used in biological processes to treat wastewater from the pulp and paper industry. Fungi degrade lignin and other phenolic compounds by producing enzymes such as lignin peroxidase laccase and manganese peroxidase. The widely used fungal species are *Merulius aureus* (Malaviya and Rathore 2007a), *Fusarium sambucinum* (Malaviya and Rathore 2007a), *Rhizopus oryzae* (Nagarathnamma and Bajpai 1999), *Phanerochaete chrysosporium* (Zhang et al. 2012a, b), *Trametes pubescens* (González et al. 2010), and *Aspergillus niger* (Liu et al. 2011). *Phanerochaete chyrosporium* and some brown rot fungi are predominantly used to treat paper and pulp mill effluent. Specifically *Pleurotus sajor caju* and *Rhizopus oryzae* species can reduce COD by 74–81% from effluents of a bleached kraft *Eucalyptus globulus* after 10 days of incubation (Freitas et al. 2009). Singhal and Thakur (2009a) studied the fungal species *Emericella nidulans var. nidulans* which was found to degrade 37% of lignin at optimum temperature (30–35 °C), pH (5), rpm (125), and inoculation size (7.5%) after 24 h. A consortium of *Merulius aureus* syn *Phlebia* sp. and *Fusarium sambucinum* Fuckel MTC3788 was studied to treat the paper mill effluent by P. Malaviya et al. (2007a) that resulted in a reduction of color, lignin, and COD by 78.6%, 79%, and 89.4% within 4 days. About 50–53% reduction in color, 35–40% lignin concentration at optimum temperature (30–35 °C), pH (5), rpm (125), and inoculation size (7.5%), after 24 h was shown by

Table 1 Various microorganisms and their lignin degradation potentials: (a) fungal species and (b) bacterial strains

(a)

Sl. no	Fungal species	Source of lignin	% lignin degradation	Incubation time	Reference
1	*Merulius aureus* syn *Phlebia* sp. and *Fusarium sambucinum* Fuckel MTC3788	Paper and pulp mill effluent	79%	4 days	Malaviya and Rathore (2007a)
2	*Emericella nidulans* var. nidulans	Pulp and paper mill effluent	37%	1 day	Singhal and Thakur (2009a)
3	*Cryptococcus* sp.	Pulp and paper mill effluent	35–40%	1 day	Singhal and Thakur (2009b)
4	*Paraconiothyrium variabile* Damm	Decay wood of *Salix matsudana* Koidz	22.99%	40 days	Gao et al. (2011)
5	*Phanerochaete chyrosporium*	Rice straw	64.25%	28 days	Zhang et al. (2012a, b)
6	*Trabulsiella guamensis*	Alkyl lignin	66%	28 days	Sumathi and Hung (2006)
7	*Aspergillus flavus*; *Emericella nidulans*	Alkali lignin	19–41.6%	21 days	Barapatre and Jha (2017)
8	*Ganoderma lucidum* BEOFB 435	Wheat straw	58.5%	21 days	Ćilerdžić et al. (2017)
9	*Ceriporiopsis subvermispora*	Wheat straw	66%	49 days	Erven et al. (2018)
10	*Myrothecium verrucaria*	Corn stover	36.73	96 h	Su et al. (2018)
11	*Shinella* sp., *Cupriavidus* sp., and *Bosea* sp.	Wooden antiques	54%	2 days	Zhang et al. (2021)
12	*Trametes hirsuta* BYL-3, *Trametes versicolor* BYL-7, and *Trametes hirsuta* BYL-8	Rice straw	39.7%	10 days	Wang et al. (2022)

(b)

Sl. no	Bacterial species	Source of lignin	% lignin degradation	Incubation time	Reference
1	*Bacillus* sp.	Paper and pulp mill effluent	53%	6 days	Raj et al. (2007)
2	*Paenibacillus* sp. strain LD1	Paper and pulp mill effluent	54%	6 days	A. Raj et al. (2014)
3	*Bacillus magnetrium* and *Pseudomonas plecoglossicida*	Black liquor	82%	7 days	R. Paliwal et al. (2015)
4	*Bacillus subtilis* and *Klebsiella pneumonia*	Kraft lignin	58%	6 days	S. Yadav and Chandra (2015)
5	*Serratia liquefaciens*	Paper and pulp mill effluent	58%	6 days	I. Haq et al. (2016)
6	*Bacillus subtilis*, *Bacillus endo-phyticus*, *Bacillus* sp.	Paper and pulp mill effluent	40.19%	2 days	Ojha and Tiwari (2016)
7	*Bacillus ligniniphilus* L1	Alkaline lignin	38.9%	7 days	Zhu et al. (2017)
8	*Planococcus* sp. TRC1	Paper mill sludge	74%	2.5 days	Majumdar et al. (2019b, a)
9	*Serratia marcescens* NITD-PER1	Paper mill sludge	54%	3 days	Majumdar et al. (2020a, b, c)
10	*Bacillus flexus* RMWW II	Alkali lignin	85%	6 days	Kumar et al. (2019)
11	*Bacillus amyloliquefaciens* SL-7	Tobacco straw	28.55%	15 days	Mei et al. (2020)
12	*Bacillus altitudinis* SL7	Paper and pulp effluent	44%	5 days	Khan et al. (2022)

Cryptococcus sp. as studied by A. Singhal et al. (2009b). Effectively 61.9% reduction in lignin has been shown by *Aspergillus flavus* strain F10 (Barapatre and Jha 2016). A consortium of *Nigrospora* sp. LDF00204 and *Curvularia lunata* LDF21 showed 80% reduction in COD and 76.1% in lignin concentration (Rajwar et al. 2017). *Bjerkandera adusta* and *Phanerochaete chyrosporium* showed the ability to completely delignify paper mill effluent within 8 days (Costa et al. 2017). *Ganoderma lucidum* BEOFB 435, *Ganoderma applanatum* BEOFB 411, *Pleurotus eryngii* HAI 711, *Pleurotus pulmonarius* HAI 573, *Pleurotus eryngii*, *Pleurotus ostreatus*, and *Pleurotus pulmonarius* have been reported to have excellent delignification capacity due to their ability to produce ligninolytic enzymes (Stajić et al. 2006; Ćilerdžić et al. 2016, 2017). Few ascomycetes such as *Podospora anserina*, *Chrysonilia sitophila*, *Chaetomium globosum*, *Ustulina deusta*, and *Alternaria alternata* have also been reported to have delignification potentiality (Ferraz and Durán 1995; Sigoillot et al. 2012; Ferrari et al. 2021). However, it has been found from several studies that fungal treatment has a few limitations, including the requirement for exogenous carbon sources, adjustment of low pH, the requirement for hydraulic retention time, and penetration of mycelium into substrates. R. Priyadarshinee et al. (2016) critically reviewed these limitations and concluded that these are creating hindrances in the commercial application of fungi in the treatment of effluents.

Bacterial treatment strategies

Bacterial strains have emerged as prospective substitutes after researching the drawbacks of using fungal species in this wastewater treatment. According to research, *Pseudomonas*, *Nocardia*, and *Corynebacterium* strains could grow fast on phenols linked to lignin but could not break down lignin (Janshekar et al. 1982). However, several research studies have reported on certain bacteria's delignification abilities, such as *Pseudomonas*, *Arthrobacterium*, and *Xanthomonas*. According to Raj et al. (2007), bacteria may absorb K.L. from paper mill effluents as the only carbon source and decompose it up to 98% of the same. Using three bacterial isolates, *Bacillus subtilis*, *Bacillus endo-phyticus*, and *Bacillus* sp., Ojha and Tiwari (2016) studied the degradation of lignin in paper industry wastewater. Their results showed 17.8%, 17.9%, and 16.87% delignification, respectively. Interestingly, when a consortium made up of these bacterial strains was applied, lignin degradation increased to 40.19%. With the use *of Bacillus megaterium*, *Bacillus subtilis*, *Bacillus* sp., and *Pseudomonas aeruginosa* in batch study experiments, lignin breakdown and color reduction from effluent from paper and pulp mills have been accomplished up to 50–97% (Raj et al., 2007; Tiku et al. 2010; Tyagi et al. 2014). Bacteria can break down only monomeric lignin, according to several findings (Priyadarshinee et al. 2016; Majumdar et al. 2020a, b, c), and only a small number of strains have been reported that can break down the complex derivatives of lignin produced during pulping operations (Chandra and Bharagava 2013; Priyadarshinee et al. 2016). Using *Serratia liquefaciens* and *Paenibacillus* sp., it has been demonstrated that lignin peroxidase and laccase induction may also be used to bioremediate paper and pulp effluent (Raj et al. 2014; Haq et al. 2016).

Bacterial treatment is gaining more interest as several bacterial strains can survive during a broad range of temperature, pH, and in the presence of toxic components. The bacterial growth rate is also faster than fungal. In the case of bacterial treatment, there is no additional cost for removing mycelium (Priyadarshinee et al. 2016). Bacterial species such as *Bacillus* sp. CS1 (Chang et al. 2014), *Sphingobacterium* sp. (Wang et al. 2018), *Comamonas* sp. B9 (Zhang et al. 2012a, b), *Pandoraea* sp. ISTKB (Kumar et al. 2015a, b), *Pandoraea* sp. B-6 (Fang et al. 2018), and *Novosphingobium* sp. B-7 (Chen et al. 2012) is gaining popularity in lignin degradation over fungal species. *Paenibacillus* sp. strain LD1 could reduce COD by 78% and lignin by 54%, as studied by Raj et al. (2014). *Serratia liquefaciens* remarkably reduce COD by 85% and lignin by 58% from paper mill effluent, as reported by Haq et al. (2016). Consortium culture of *Bacillus magnetrium* and *Pseudomonas plecoglossicida* immobilized on corn cob agricultural residue showed remarkable efficiency in biodegradation of black liquor by mitigating 79 and 82% of COD and lignin, respectively, after 168 h of bacterial treatment (Paliwal et al. 2015). *Pseudomonas* holds a significant position in wastewater remediation processes due to its remarkable biodegradation capability of several persistent xenobiotic compounds (Zhu et al. 2015). Recently *Paenibacillus gluanolyticus* showed the potential to deconstruct lignin found in black liquor into value-added products like ethanol, succinic, lactic, propanoic, and malonic acid (Mathews et al. 2016). Among other bacterial species, *Bacillus flexus* RMWWII effectively reduced the alkali lignin concentration found in the effluent (Kumar et al. 2019). *Bacillus cereus* AKRC03 could decolorize and degrade 78.67% of hazardous organic pollutants of paper mill effluent (Kumar and Chandra 2021). Recent studies by Khan et al. (2022) showed that *Bacillus altitudinis* SL7 could degrade 44% lignin if a load of lignin in the effluent is below 3 g/l.

Apart from the various bacterial species studied, our laboratory found a less explored bacterial species from the genus *Planococcus* showing delignification capacity. The specific role of *Planococcus* sp. TRC1 for the abatement of paper mill effluent has been investigated (Majumdar et al. 2019b, a). The results revealed a reduction in 85% COD, 74% lignin, 81% phenol, and 96% color after 60 h of treatment. In this unique strategy, the bacterial strain was immobilized on the surface of paper mill sludge beads and used in a fluidized bed reactor to detoxify paper industry wastewater. The use of *Planococcus* in diverse biotechnological applications is steadily gaining momentum (Behera et al. 2010; Satpute et al. 2010; Engelhardt et al. 2001). Li et al. (2006) reported 100% removal of 1 mM toluene after 32 h of incubation by *Planococcus* spp. under hypersaline conditions. The catabolic ability of *Planococcus* sp. for the biodegradation of aromatic hydrocarbons under extreme conditions might be attributed to the presence of essential genes which can translate the stress response proteins, as revealed during decoding the first whole-genome sequence of *Planococcus antarcticus* DSM (Margolles et al. in 2012). Although several investigations on the degradation of complex hydrocarbons by *Planococcus* species have been performed (Satpute et al. 2010; Engelhardt et al. 2001), seldom the ligninolytic activity of this potential species has been explored; however, KL degradation efficiency of this genus has been studied by our group.

Biological treatment of PMS

Looking at environmental safety and effectiveness, biological treatment is gaining importance for the solid waste also. With delignifying microorganisms like ascomycetes, basidiomycetes, bacteria, and enzymes, deconstruction of PMS is possible (Zeng et al. 2012). As a result, delignified PMS achieves more exposure for enzyme digestion of cellulosic fraction (Karlsson et al. 2011). It has been found that brown rot and soft rot fungi attack the cellulose material of lignocellulose and can only impart a minor impact on the lignin fraction (Priyadarshinee et al. 2016). So comparatively, white rot fungi has become the microbe of choice, which degrade the lignin component more actively than cellulose (Zeng et al. 2012). In this context, bacterial strains are not way behind; strains of *Planococcus* sp. TRC1, *Pseudomonas fluorescens* NITDPY, and *Serratia marcescens* NITDPER1 have shown the potential of selective lignin deconstruction from biomasses of kraft pulp and PMS, respectively, which can be significant candidates of future choice in pulp and paper industries (Priyadarshinee et al. 2015; Majumdar et al. 2020b). *Bacillus* sp. IITRDVM-5 showed tremendous potential for remediation of PMS (Sonkar et al. 2021). Biological pretreatment has several advantages like no chemical requirement, lesser energy requirement, and is environment-friendly.

Waste treatment and recovery of valuables: a key platform

Apart from protecting the environment from undesired waste impacts, wastewater treatment is a crucial platform for valuables recovery. There is demand for some new generation technology for wastewater treatment plants, where energy, organics, and other resources can be recuperated as valuable by-products instead of being wastefully destroyed. Recycling waste will significantly support the scheme of product diversity and the power of economic growth (Landrigan and Fuller 2015). Currently, researchers and industries are focusing on the integrated biorefinery approach to protect the environment and dig out cost–benefit from this waste. Figure 3 represents the biorefinery approach for pulp and paper mill waste. In industry, two routes can be implemented for the biorefinery approach. The first one is the presence of integrated biorefinery machinery at the same establishment of the industry. The second route involves collecting effluent from industries at a definite place and extracting valuables via various approaches. The second model may create a new industrial sector for the usage of waste generated by other industries. However, in the case of the second model, the cost of transportation may increase capital expenditure (Sailwal et al. 2020).

Although the composition of PMS greatly varies among paper mills depending on the raw materials used, it is majorly lignocellulosic biomass (Sonowal et al. 2014). Various studies have revealed the potential applications of PMS in numerous ways, including the production of biogas (Lopes et al. 2018), pyrolysis (Cho et al. 2017), ethanol, compost production (Boshoff et al. 2016; Hazarika et al. 2017), material preparation (Goel and Kalamdhad 2017), and vermicomposting (Negi and Suthar 2018). Apart from these functions, recently, Majumdar et al. reported unique properties of PMS such as acting as a carrier matrix for bacterial species in solid state fermentation (Majumdar et al. 2019a, b), a substrate for bacterial pigment production (Majumdar et al. 2020a, 2020b), and source of cellulose nanocrystals through bacterial delignification (Majumdar et al. 2020b). Due to its lignin content, it can be thought of as a source of lignin-based valuables such as lignin nanoparticles, as a binder for particleboard and similar laminated or composite wood products, as a soil conditioner, as a filler or an active ingredient of phenolic resins, and as an adhesive for linoleum, source of vanillin (synthetic vanilla) and dimethyl sulfoxide. Similarly, it can act as a low-cost source of cellulose-based valuables like cellulose microcrystals, carbon dots, and carbon nanotubes that have gained immense industrial importance recently (Veluchamy and Kalamdhad 2017a, b, c, d). Table 2 represents the different valuable compounds generated from PMS from industrial perspectives through chemical, physical, or biological treatment methods.

Lignin: the untapped wealth from paper pulp wastewater and sludge

Wastewater and PMS from paper industries contain lignin, hemicellulose, phenol, and some common xenobiotics (Strassberger et al. 2014). Usually, cellulose and hemicellulose are widely used to produce sugar, paper, and biofuels, while lignin is yielded from these industries (Cao et al. 2018). As a result, lignin remains the most underrated component of lignocellulosic biomass due to its high calorific value and use as an energy provider in the pulp and paper industry (Banu et al. 2022). Figure 4 represents the various valuables that can be generated from lignin isolated from wastewater or sludge. Traditionally, lignin is used for the pulp and paper industry's bioenergy generation for the mill. It is the primary producer of lignin to generate energy for running the mill and produce paper and additional lignin. Kraft lignin (KL) is the primary energy source in kraft mills, whereas lignosulfonates are commercialized for various applications (Dessbesell et al. 2020). However, recently greater interest of scientists and researchers in lignin because of its excellent properties has paved the way for discoveries and innovations. Extensive studies

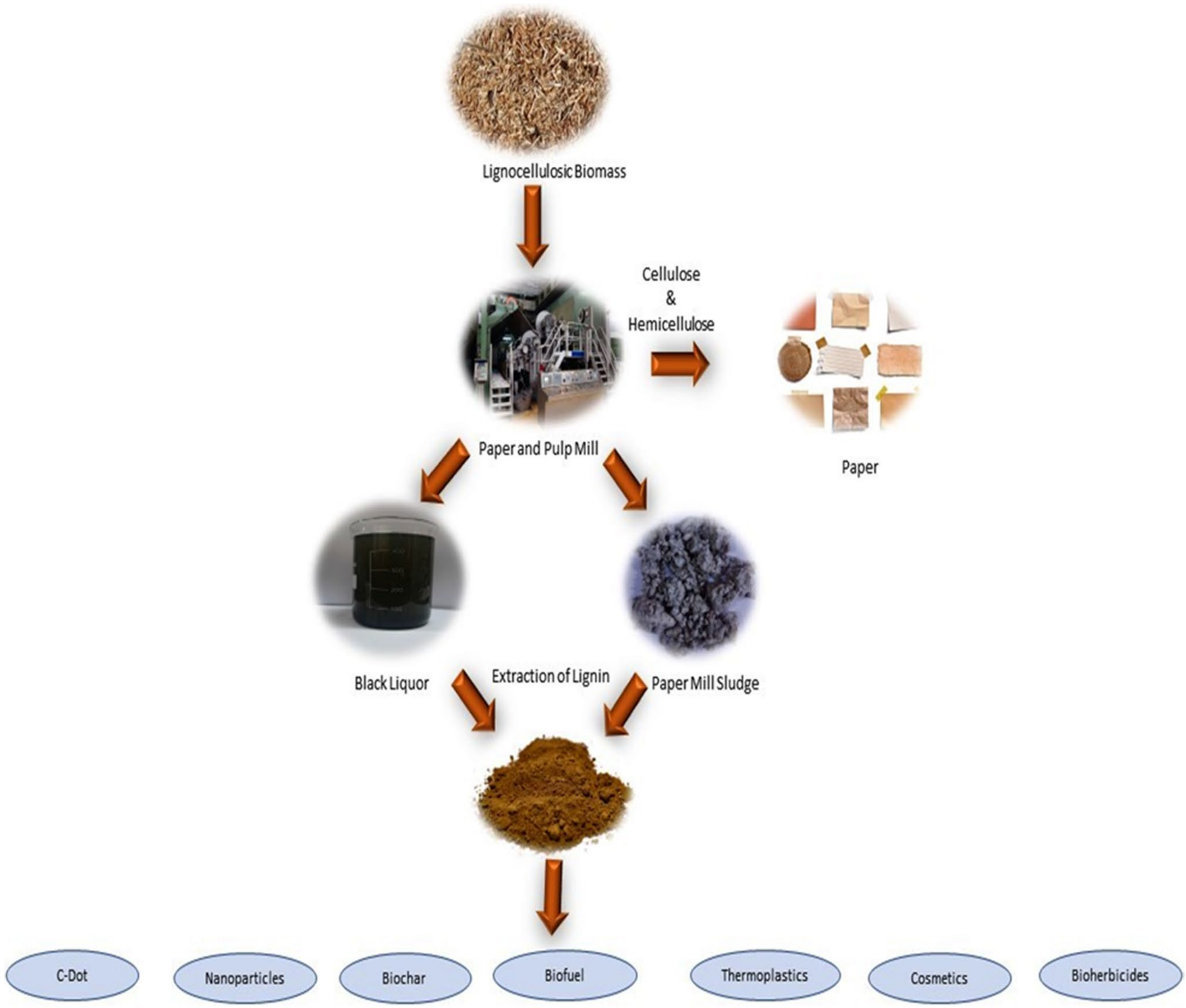

Fig. 3 Extraction and utilization of lignin from pulp and paper mill industries

are being done, and more new and novel techniques are being developed. However, it is scarce compared to other biopolymers. One of the main obstacles to developing novel products from lignin is its complex structure and valorization methods. The recalcitrant nature of lignin is a more significant caveat to producing valuable and novel products (Domínguez-Robles et al. 2020; Banu et al. 2022). Table 3 represents the various valuable metabolites generated from diverse lignin sources through biological methods. By valorizing this technical lignin, they can be used for many different purposes (Banu et al. 2022).

Utilization of black liquor for recovery of lignin

Lignin isolation from spent or black liquor is a process that has been in operation since the beginning of the twentieth century. The black liquor that contains approximately 35–45% lignin can be utilized to produce energy and several chemical commodities (Zhu et al. 2015). It can be utilized for various value-added product generations, but its use is still limited. Extraction of lignin from black liquor can be a tedious job. However, researchers have come up with various chemical procedures such as acid precipitation, ultrafiltration, and ion exchange to extract lignin from black liquor (Zhu et al. 2015; Oliveira et al. 2020).

The purified lignin can be directly converted into activated carbon, phenol, bio-oil, aromatic compounds, and other exceptional value-added products (Cao et al. 2018). There are different uses of black liquor. Roy et al. (2019) reported that when it is heated to 400–900 °C in anoxic conditions, it can be converted into biofuel, syngas, and biochar. Syngas and biofuels can be utilized to produce energy. In

Table 2 Valuables generated from PMS through various treatment methods

Treatment method	Valuable generated	Uses	References
Chemical	Activated carbon and bioactive adsorbent	Solvent recovery, gas refining, air purification, exhaust desulfurization, and deodorization processes	Khalili et al. (2002)
Chemical	Manufacture of fired bricks	High durability bricks	Goel and Kalamdhad (2017)
Mechanical (microfluidization)	Cellulose nanofibrils	Reinforcing nanofillers, biomedicine, optoelectronic devices, sensors, and energy storage devices	Du et al. (2020)
Chemical	Sintered ceramics	Flame tubes, firing trays etc	Asquini et al. (2008)
Supercritical water treatment	Energy generation	Unconventional energy source	Zhang et al. (2010)
Chemical	Wood plastic composite	Composite material	Soucy et al. (2014)
Biological	Bioethanol	Biofuel	Kang et al. (2010)
Biological	Beta-carotene pigment	Drug, food colorant	Majumdar et al. (2020a)
Biological	Prodigiosin pigment and cellulose nanocrystals	Drug, food colorant, drug delivery system, filler	Majumdar et al. (2020b)
Biological	Biogas	Energy generation	Priadi et al. (2014)
Biological	Vermicompost	Green fertilizer	Negi and Suthar (2018)

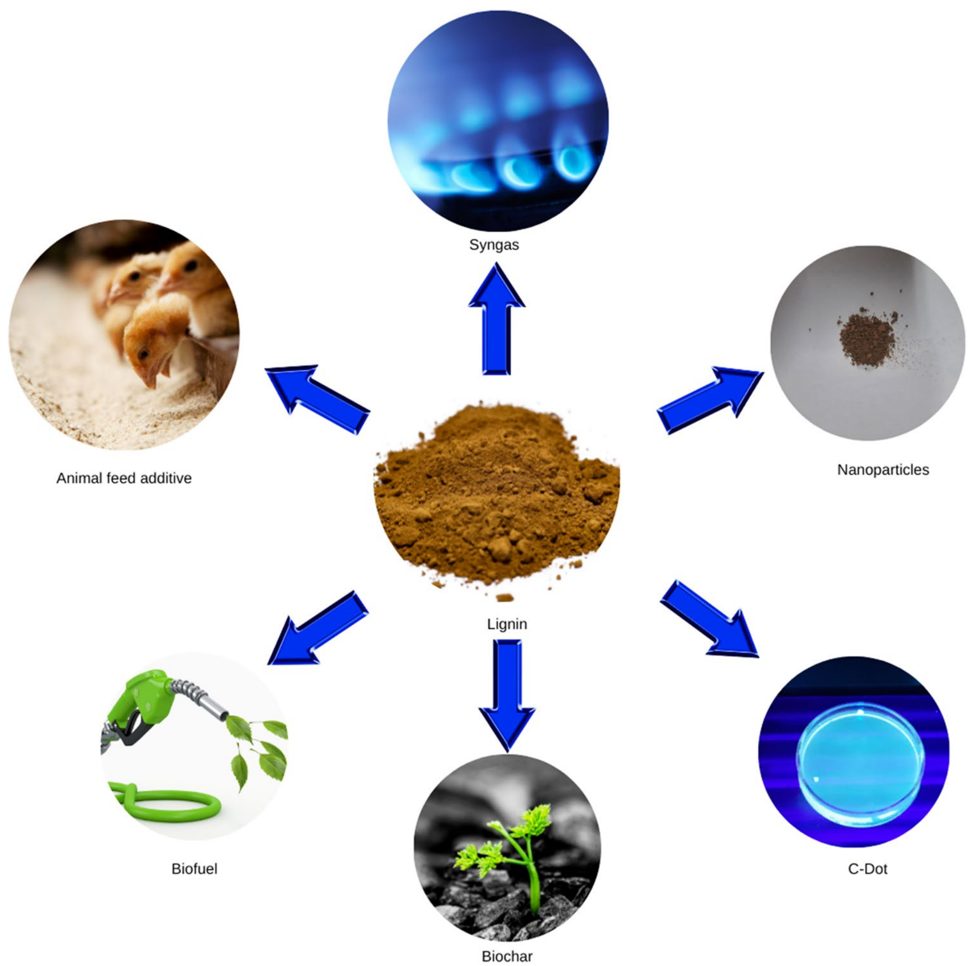

Fig. 4 Utilization of lignin extracted from pulp and paper mill waste into various valuables

contrast, biochar can be used as biofertilizers, adsorbing agents, sequestering environmental CO2, and immobilizing heavy metals such as arsenic and cadmium in the environment (Roy et al. 2019; Sailwal et al. 2020). Recently *Paenibacillus gluanolyticus* showed the potential to deconstruct lignin found in black liquor into different value-added products like ethanol, succinic, lactic, propanoic, and malonic acid (Mathews et al. 2016).

Usage of lignin in specialized valuables

The lignin valorization rate of annually produced lignin is less than 2%, proposing the need for technological improvement to get benefit from lignin as a versatile feedstock (Dessbesell et al. 2020; Cao et al. 2018). In recent years, efficient utilization of lignin has attracted wide attention. Studies indicate advancements in two significant lignin methodologies utilization: catalytic degradation into aromatics and thermochemical treatment for carbon material production. Hydrogenolysis, direct pyrolysis, hydrothermal liquefaction, and hydrothermal carbonization of lignin are some of the processes that are being explored (Dessbesell et al. 2020; Cao et al. 2018). Various value-added products could be produced from lignin, such as biomedical materials, copolymer composites, resins, antioxidants, adsorbents, catalysts and catalyst support, and carbon electrodes (Kai et al. 2016; Chen et al. 2018). In addition, many biodegradable polymers like polylactic acid (PLA), polyhydroxyalkanoates, polyhydroxybutyrate and non-biodegradable polyolefin (PL), polyurethane (PU), polyester (PE), polyol (PO), and polyamide (PA) are produced using lignin (Banu et al. 2022). Also, many other value-added chemicals and materials like important intermediate for organic synthesis—ethylbenzene, cholagogue drug material—hydroxyl acetophenone, and carbon-based supercapacitor and catalyst are made from lignin (Cao et al. 2018).

In biomedical and pharmaceuticals, lignin is a relatively new biopolymer. However, recent studies have revealed its potential for being one of the most helpful biopolymers. It has proven to elicit many health benefits due to its specific properties like antimicrobial, anti-inflammatory, anti-carcinogenic, prebiotic, antioxidant (Ayyachamy et al. 2013; Cheng et al. 2020), susceptibility to enzymatic degradation (Culebras et al. 2021), and lower cytotoxicity. These properties make it one of the potential resources for developing many novel systems for biomedical and pharmaceutical applications. In recent times, some of the most sought-after developments in these regards are hydrogels, drug delivery system platforms, nanoparticles, nanotubes, oral solid dosage forms, sunscreen lotions, wound dressing, wound healing material, pharmaceutical excipients, tissue engineering materials, gene delivery systems (Domínguez-Robles et al. 2020), bionic sensor or materials, and UV blocking material (Culebras, et al. 2021). Lignin is widely studied and used in various industries (Haq et al. 2020), such as fuels, agriculture, and construction materials (Ayyachamy et al. 2013). Interestingly, lignin is used for energy generation as an alternative fuel for power and heat generation in industries, as surfactants, resins, bonding agents, and dispersants for ceramics and concrete, polyurethane (PU) foams, vanillin production, and animal feed additive (Dessbesell et al. 2020; Cao et al. 2018). Apart from these direct uses of lignin, two major lignin-derived components of enormous industrial importance, lignin nanoparticles and carbon dots can be considered as next-generation valuables from waste.

Lignin nanoparticles

One of the most lucrative products derived using lignin in today's biomedical world is lignin nanoparticles. Biopolymeric nanoparticles generally have favorable dispersibility, high specific surface area, flexible molecular design (Dai et al. 2017), high drug loading capacity, prolonged half-life in the bloodstream, and sustained drug release behavior (Liu et al. 2018a, b). They can optimize the pharmacokinetics and pharmacodynamics of drugs (Dai et al. 2017). Bio-based nanomaterials and nanoparticles most commonly used as drug delivery systems are polymer–drug conjugates and liposomes (Cheng et al. 2020). LNPs have improved antioxidant and UV blocking properties compared to macromolecular lignin particles (Domínguez-Robles et al. 2020). Lignin nanoparticles have great potential as nano and micro-carriers of soluble drugs, pesticides, genes, and food additives (Venkatesan et al. 2009; Tsuji 2001; Green et al. 2006; Desai et al. 2005). It can be used as a surfactant in Pickering emulsions. Reports showed that lignin nanoparticles of about 320 nm made from kraft lignin have effectively stabilized hexadecane droplets in aqueous emulsions (Nypelö et al. 2015). Lignin has remarkable free radical scavenging properties, reducing oxygen radicals; therefore, it can be a powerful antioxidant (Lu et al. 1998). Moreover, Zimniewska et al. (2008) showed that fabrics treated with nanostructured lignin possess excellent UV resistance.

Nanoparticles synthesized from biopolymer have been widely used in drug delivery systems or RNA-interfering effectors (Yearla and Padmasree 2016). Lignin nanoparticles effectively improve membranes' mechanical properties as cellulose triacetate (Dia et al. 2017). Dai et al. (2017) reported efficient delivery of resveratrol. Besides, they contain different monomers with an antibacterial activity that cause cell membrane damage and bacterial cell lysis and can even act as a natural biocide (Cazacu et al. 2013). Extraction of lignin from pulp and paper industry wastewater

Table 3 Various valuable metabolites generated from diverse lignin sources through biological methods

Degradation method	Bacterial/fungal sp. used	Material used	Metabolites formed	Reference
Bacterial degradation	*Paenibacillus* sp., *Aneurinibacillus aneurinilyticus*, and *Bacillus* sp.	Kraft lignin	Acetic acid, gallic acid, propanoic acid, guaiacol, valeric acid, ferulic acid, *t*-cinnamic acid, bis(2-ethylhexyl) phthalate	A. Raj et al. (2007)
Bacterial treatment	*Novosphingobium* sp. B-7	Bamboo slips	Octadecanoic acid, hexadecenoic acid, 3-methoxy-4,β-dihydroxy benzene propanoic acid, vanillic acid, butylated hydroxytoluene, salicylic acid, acetylsalicylic acid, lactic acid, 3-methyl-2-butanol, 3,5-dimethyl benzaldehyde	Y. Chen et al. (2012)
Bacterial and fungal	*C. versicolor*, *T. gallica*, *Mycobacterium* sp., *Streptomyces* sp.	Kraft lignin	Apocynin, vanillic acid, guaiacol	F. Asina et al. (2016)
Bacterial and fungal degradation	Mixture of various strains including Ascomycota, Glomeromycota, Proteobacteria, Bacteroidetes, Actinobacteria	Oil palm empty fruit bunches	Catechol, benzoic acid, apocynin, vanillin	Tahir et al. (2019)
Bacterial treatment	*Streptomyces* sp. S6	Kraft lignin	Triphenylphosphine oxide, bis(2-ethylhexyl) phthalate, butyl acetate, 3-methyl-butanoic acid, 2-methoxyphenol (guaiacol)	Riyadi et al. (2020)
Bacterial treatment	*Bacillus cereus*	Paper and pulp wastewater	Methyl isoleucine, hexadecane, pregna-3,5-dien-20a-ol, O-trimethylsilyl, 4 methyl-10-(1-phenyl-ethylidine)-2,4,6-triaza-tricyclo [5.2.1.0(2,6)] decane	R. Kumar et al. (2022)

can lead to a significant source for producing these wonder molecules that cannot only pave the way toward a waste-to-wealth approach but also be an excellent tool for mitigating toxic wastewater from paper industries.

In recent decades due to the extensive research and developments in nanoscience and nanotechnological studies, their implication in the development of nanomedicine and pharmaceuticals has just exploded. It has become one of the hot topics in the research community. Many lignin-based novel nanomaterials and biopolymers have already been developed. It is sometimes regarded as the most up-and-coming technology in the future as a way to overcome medical problems (Cheng et al. 2020) due to the magnetic properties of polymer-based nanoparticles. Biopolymeric nanoparticles generally have favorable dispersibility, high specific surface area, flexible molecular design (Dai et al. 2017), high drug loading capacity, prolonged half-life in the bloodstream, and sustained drug release behavior (Liu et al. 2018a, b). They can optimize the pharmacokinetics and pharmacodynamics of drugs (Dai et al. 2017). Lignin has other use as well due to its excellent properties. Some of these less explored fields are the use of lignin as a pharmaceutical excipient and other materials. Gao et al. (2021) created lignin-based fluoroquinolone antibiotics removing adsorbents by using actinia-shaped lignin-based adsorbents (LNAEs), using lignin as core, and designing a tentacle of grafted poly-acrylic acid (PAA). They were used to adsorb ofloxacin and ciprofloxacin from water. One such sunscreen was developed by Lee et al. (2020). Light-colored lignin (CEL) was extracted from rice husks by cellulolytic enzyme treatment and solvent extraction then spherical nanoparticles (CEL-NP) were prepared using a solvent shifting method. They exhibited higher SPF and UVA PF. Other uses of lignin-based biopolymers include gene delivery systems (Domínguez-Robles et al. 2020) and other precursor molecules. Table 4 lists various lignin-based products with biomedical and pharmaceutical applications.

Bio-based nanomaterials and nanoparticles most commonly used as drug delivery systems are polymer–drug conjugates and liposomes (Cheng et al. 2020). LNPs have improved antioxidant and UV blocking properties compared to macromolecular lignin particles (Domínguez-Robles et al. 2020). Cheng et al. (2020) developed intelligent lignin-based self-assembling nanomicelles for oral drug delivery. The nanomicelles were pH-responsive and cell culture showed the ability to inhibit the survival of human colon cancer cells HT-29 while proliferating human bone marrow stromal cells hBMSC and mouse embryonic fibroblast cells NIH-3T3. Ibuprofen (IBU) was used as a model drug. The lignin nanoparticles have great potential in cancer treatment and drug delivery. About 40– 60% of the new drugs are generally hydrophobic (Domínguez-Robles et al. 2019), whereas drug carriers like lignin hydrogels are bad at loading and delivering hydrophobic drugs. This problem is mainly solved by the nanoparticles and nanomicelles developed using lignin and other amphiphilic polymers (Cheng et al. 2020).

Recently three-dimensional-printed biomaterials have become a popular study topic for developing excellent biomedical and pharmaceutical products. 3d-printed wound dressing is one of them. Lignin and antibiotic (tetracycline) are combined with poly(lactic acid) (PLA) pellets, coated with castor oil and polyhydroxybutyrate (PHB) to create

Table 4 Lignin-based pharmaceuticals and biomedical techniques and products

Technique/methodology	Type	Mechanism and findings	References
Hydrogel-based drug delivery system for controlled release of paracetamol	Drug delivery system	Hydrogel prepared using crosslinking poly(ethylene) glycol diglycidyl ether (PEGDGE) for delivering paracetamol. It is found that greater the crosslinking density greater the hydrogel swelling and more hydrophilic groups yielding more control over delivery and pore size. Lignin:PEGDGE 1:1 ratio showed higher drug diffusion for paracetamol	Culebras et al. (2021)
Hydrogel-based drug (curcumin) delivery system	Drug delivery system	Hydrogels were prepared from lignin by combining lignin with poly(ethylene glycol)/PEG and poly(methyl vinyl ether-co-maleic acid) by esterification reaction Curcumin was used as model drug for controlled delivery for up to 4 days	Larrañeta et al. (2018)
Stretchable, conductive, self-healing, and ultraviolet-blocking Fe-SL-g PAA hydrogels	Potential bioelectric and bionic sensor	Fe-SL-g-polyacrylic acid (PAA) hydrogel was developed in rapid timescale from sulfonated lignin (SL) and $Fe3+$ at 20 °C by dynamic redox reactions with APS	Wang et al. (2020)
Lignin-based self-assembling	Drug delivery system	Lignin-based self-assembling nanomicelle was prepared by synthesizing macromonomer	Cheng et al. (2020)

such materials which can be used as 3d printing biocomposite filaments (Domínguez-Robles et al. 2020; Yu and Kim 2020). As a result, wound dressing mesh could be developed using 3d printing technology with the extra benefit of customization in size and shape. Another potential use of 3d-printed biomaterials is the fabrication of precise scaffolds for biomedical and tissue engineering applications. Zhang et al. (2020) developed spherical colloidal lignin particles and cellulose nanofibril-alginate hydrogel-based 3d-printed scaffold by combining cellulose nanofibril hydrogels with alginate through crosslinking in the presence of Ca2 + ions. These particles are known as spherical colloidal lignin particles (CLPs). These CLPs were combined with CNF-alginate to create cellulose nanofiber (CNF)-alginate-CLP nanocomposite scaffolds. As noticed, they were found to be biocompatible and thus can be used to print scaffolds for soft-tissue engineering and regenerative-medicine applications.

Carbon dots

Nowadays, a new type of carbon-based nanomaterial, carbon dots (CDs), has attracted broad research interest because of their various physicochemical properties and favorable features like good biocompatibility, unique optical properties, low cost, eco-friendliness, abundant functional groups (e.g., amino, hydroxyl, carboxyl), high stability, and electron mobility (Chen et al. 2016). Carbon dots (CDs) are a type of fluorescent carbon nanomaterial with several distinct characteristics, including tunable emission, good biocompatibility, low cost, and ease of manufacture. They have been used in cellular imaging, sensors, drug delivery, solar cells, and catalysis, among other things (Lim et al. 2015; Wang et al. 2017). They are very flexible materials and can be quickly fixed in sturdy holders for chemical and biochemical testing (Raj et al. 2007).

Sun et al. (2021) synthesized hydrogel containing lignin-based carbon dots. Liu et al. (2020) showed the formation of luminescent transparent wood based on lignin-derived carbon dots as a building material for dual-channel, real-time, and visual detection of formaldehyde gas. Rai et al. (2017) and Myint et al. (2018) prepared lignin-derived carbon dots with excellent bioimaging properties. Researchers have revealed the synthesis of carbon dot (CD), an exciting type of carbon nanoparticle with high luminescence by hydrothermal treatment of lignin in the presence of H_2O_2 that can be useful as bio-imaging sensors for bioanalytical diagnostics (Chen et al. 2016). CD can be utilized for various other applications, such as oxygen reduction reaction (Mohideen et al. 2020), as the catalyst for green oxidation of phenol (Pirsaheb et al. 2018), pH sensor, moisture sensor, solvatochromism, and solid-state multicolor lighting (Moniruzzaman and Kim 2019). But such application of lignin for the production of CD is still scarce in the literature, and the raw materials used for carbon dots preparation are restricted to lignocellulosic biomasses and other carbon-based natural substances. In this regard, lignin extracted from pulp and paper industry wastewater may be a valuable source for the same generation. It will reduce the cost of producing these CDs and lead to unleashing a new niche for utilizing lignin from waste.

Utilization of PMS through microbial treatment for valuable compounds

Microbes' role in valorizing waste has always been critical to industries. These organisms not only play a crucial role in biodegradation but also are environmentally safe and acceptability is higher. Several studies have utilized these organisms to valorize PMS and generate valuable compounds. Figure 5 represents the various strategies of microbial treatment for the generation of valuables from PMS. These involve technologies like anaerobic digestion for the generation of biogas. Priadi et al. (2014) reported that anaerobic digestion uses diverse groups of natural bacteria for biodegradation and aids in producing renewable energy from discarded organic material-rich waste. The biogas generated through anaerobic digestion could be used for heating the digester or lighting in the industry. But, anaerobic digestion of PMS is still in its infancy as compared to wastewater (Meyer and Edwards 2014). This is because of the recalcitrant nature of the lignocellulose content of PMS, making the hydrolysis critical. In a study by Lin et al. (2017), it was found that methane yield was increased 1.4-fold by pretreating PMS feedstock with the active bacterial consortium, and the maximum methane yield of 429.19 ml/g/S was noted.

Some recent studies have shown great prospects among other microbe-based treatments of PMS. Looking at the nutritive composition of PMS, it has been used as a growth substrate for microbes. Majumdar et al. (2020a) revealed that *Planococcus* sp. TRC1 could grow on PMS in solid-state fermentation (SSF), giving celebrated productivity of the pharmaceutically important compound β-carotene. This study revealed an 84% cost reduction of β-carotene from the current market price when PMS was used as the substrate. Besides that, the residual PMS biomass after bacterial pigment production was bioconverted into cellulose-rich biomass due to the promising lignin-degrading capacity of the microbe. Such cellulose-rich biomasses hold excellent prospects when industrial importance is concerned.

In another study by Majumdar et al. (2020b), PMS was dual valorized through the action of bacterial isolate *Serratia marcescens* NITDPER1. This isolate produced anticancer, antibiotic, and anti-inflammatory compound prodigiosin in SSF, utilizing PMS as substrate. The residual PMS biomass was a potential source of cellulose nanocrystals

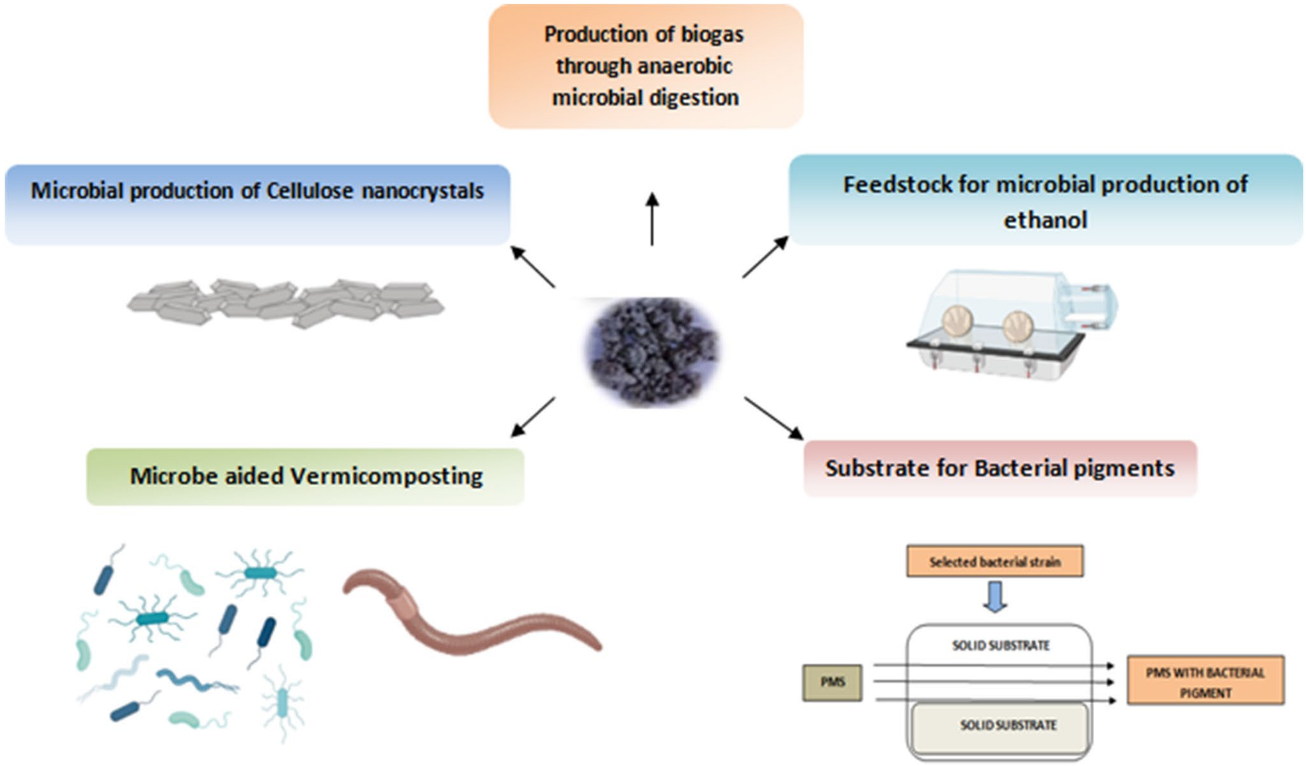

Fig. 5 Strategies of microbial treatment for the generation of valuables from PMS

with excellent thermal stability. The products generated through the processes were confirmed to be non-toxic and safe for industrial use. Such studies hold great potential in presenting PMS as the next-generation lignocellulosic biomass for cellulose and lignin-based valuables through microbial action.

Another essential utilization of PMS through microbial action has been observed during vermicomposting. In association with earthworm species, the addition of microbial species *Oligoporus placenta* in PMS has shown remarkable results in decreasing total organic carbon, C/N ratio, and cellulose but an increase in total Kjeldahl nitrogen, total phosphorus, total potassium, and pH during vermicomposting. This study by Negi and Suthar (2018) revealed that fungal inoculation during vermicomposting was effective in decomposing cellulose-rich PMS and aided in better quality compost generation.

In the studies of Kang et al. (2010), two types of paper mill sludges, primary and recycle sludge, were used as feedstock for bioconversion to ethanol. The study revealed that commercial cellulase was inefficient because of interference with ash. Simultaneous saccharification and co-fermentation (SSCF) strategies were used involving cellulase and recombinant *Escherichia coli* (ATCC-55124), and simultaneous saccharification and SSF were followed using cellulase and *Saccharomyces cerevisiae* (ATCC-200062). Celebrated ethanol yields of 75–81% were achieved from the SSCF. The work of Niju and Vijayan (2020) can also support such studies, where PMS has been demonstrated as a potential feedstock for microbial ethanol production. The various values generated from PMS through various treatment methods are represented in Table 2. These findings highlight the role microbes possess in the valorization of waste PMS for the benefit of society. Besides these, cellulolytic bacteria or enzymatic treatment may also be beneficial in extracting the lignin fraction from PMS for valorization. However, such studies are scarce in literature but hold excellent prospects for future research on PMS valorization.

Challenges, prospects, and concluding remarks

The paper industries are growing daily, and their demand is constantly increasing. Although not severe, due to the excessive amount of waste, these industries' impact on the environment is very alarming. These industries produce modified lignin, the main component of wastewater, which places the local environment vulnerable to contamination. Biological processes are beneficial because treatment must be environmentally friendly. Waste disposal is a high cost for industries and a significant concern. The significant drivers for waste valorization from the pulp and paper industry are economy and environmental expertise and industries pushing to recover and regain all these substances because 50 and 100% of lost waste resources are contained in wastewater. Recovering valuables from this waste can offset industry costs and reduce the pollution load on the local environment. This holistic approach to extracting wealth from waste must correctly identify the problems that exist in the industry. It is challenging to find out economically viable solutions for optimizing material recovery and production. Environmental impact assessment and industry life cycle assessment will help provide a new way to develop better waste utilization processes while minimizing the environmental burden. Considering the previous report, the industry's design should adopt an interdisciplinary approach, considering the theory of circular economy, green chemistry, and industrial ecology.

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• Subhasree Majumdar—contributed in literature survey on paper mill sludge related scientific information and manuscript writing

• Protik Chanda—contributed in literature survey in specific area and collection of data

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