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REVIEW ARTICLE

Current approaches and future potential for pretreatment of agricultural residues and industrial effluents to boost biomethanation

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Abstract

The burning of organic agricultural substrates such as crop residues, paddy straw, maize stalk, sugarcane straw and corn stover leads to severe air pollution. These wastes are essential feedstocks in the production of biogas. The above materials can be anaerobically digested to produce biogas, which in turn can be used to generate fuel, cooking gas, soil-conditioner and electricity as a sustainable alternative use for these residues. However, the presence of lignin and cellulose contents in these wastes, at concentrations ranging from 6-26 % and 5-50 %, reduce the effectiveness of biomethanation process. For optimum anaerobic digestion, appropriate pretreatment methods, such as physical (milling, grinding, ultrasonic), chemical (alkali, thermo-chemical pretreatment) or biological (enzymes, microorganisms) techniques, can be used to lower the lignin content. Untreated effluents containing organic matter, fertilizers, heavy metals and other contaminants are released into water bodies by agro-industries, resulting in the degradation of land and water ecosystems. Like agricultural substrates, agro-industrial effluents can be efficiently used to produce bioethanol and biogas, following the removal of these inhibitors by using effective pretreatment methods. When utilized as a feedstock, pre-treated wastewater can yield up to 2.8 % more biogas and 64 % more methane than untreated wastewater. The biodigested slurry improves soil health and enhances crop yield. This article describes several pretreatment techniques to improve biogas production from industrial effluents and agricultural wastes entrusting the soil health, sustainability and crop yield, consistent with the circular economy concept.

Keywords: biogas; enzymes; inhibitors; methane; pretreatment; soil conditioner

Introduction

Crop residues, animal manure and other organic wastes are hard to dispose of due to their heterogeneous composition and complex makeup. These agricultural wastes could be aptly pretreated and anaerobically digested to produce various second and third-generation fuels (1). Improper management of these agricultural wastes has resulted in soil, air and water pollution leading to global warming. Instead of open field burning, these wastes can be properly utilized as feedstock for the generation of biogas, bioethanol and biohydrogen. This innovative conversion ensures environmental safety and renewability of available resources (2). Agricultural wastes, vinasse, swine wastes and other animal wastes have been digested to produce methane, ensuring their safe disposal after utilization. Adopting the most efficient technology increases the quality and quantity of biogas production (3). Agricultural wastes generated can be recycled using conventional methods (biogas, composting) and non-conventional technologies (used for filtration and adsorption) to enhance environmental sustainability (4). Shortages of labour, transport and other facilities hinder the disposal of waste. These challenges can be tackled by on-farm management practices like mechanization, mulching and off-farm practices including production of biogas and composting (5). Limited availability of natural resources has led to increased use of alternative renewable energy sources like solar, wind, hydro-thermal energy, etc. Biogas production technology serves as the best method for the conversion of organic wastes into biogas, thereby improving the livelihood and ecosystem. Anaerobic fermentation of agricultural wastes and other allied biomass generates biogas containing 60 % methane and 45 % carbon dioxide (6). Biogas produced from agricultural wastes reduces both the environmental and financial problems of farmers, subsequently reducing reliance on costlier energy sources and promoting a safe and sustainable way of waste management (7). Biogas generation using these agricultural wastes as co-digestates strengthened the concept of a circular economy (8). Though lignocellulosic materials are the best substrates for anaerobic digestion, they impede biotransformation, becoming the rate-limiting step in the process (9). In India, about 500 -550 Mt of crop residues are generated annually. Among these, wheat (110Mt), rice (122Mt), maize (71 Mt), sugarcane (141Mt) and pulses (28Mt) are the major contributors. Rather than being disposed of in the environment, these residues could be efficiently utilized for the production of bioethanol or biogas (10).

India generates about 61754 million litres per day (MLD) of industrial effluents, of which only 22963 MLD is treated, while the remaining 38791 MLD is discharged into the aquatic ecosystems, causing severe pollution and health hazards (11). The discharge of untreated industrial effluents into the environment causes various health issues, like cancer, as well as skin and liver ailments. Thus, the effluents can be treated and used as feedstock for the production of second-generation fuels (12). Integration of biomethane production using industrial effluents as feedstock ensures their safe discharge while enabling the regeneration of renewable energy like biogas, biohydrogen, bioethanol, etc. (13). Biomethanation using industrial effluents is impeded by the presence of phenols, antibiotics, organic acids, ammonia, sulfides, phosphorous, heavy metals, aliphatic nitrogen compounds, etc. Pretreatment of these effluents before anaerobic digestion enhances the microbial population and biogas production (14). The concentrations of ammonia above 4 gL^{-1}, volatile fatty acids above 28 $g\ L^{-1}$ and phenolic acids above 4 gL^{-1} inhibit microbial load and methane production (15, 16). To mitigate the effects of these inhibitors, strategies like microbial acclimation, inoculum enrichment, chemical, physical adsorption and effluent dilution can be adopted (17). The biomethanation process begins after ammonia volatilization, followed by the removal of potassium and phosphorous. This process also depends on the microbes involved in the fermentation (18). Industrial effluents, a rich source of inorganic and organic toxic pollutants, pose a great threat to environmental sustainability. These pollutants are carcinogenic, mutagenic and teratogenic, affecting human health and soil fertility (19). Unwanted wastewater obtained from industries can be treated physically, chemically or biologically, after which the pretreated water is used for fertigation. Being rich in nutrients, the treated water helps reduce fertilizer cost and water scarcity issues. It also reduces the effect of soil and water pollution, aiding in the production of biogas and in the creation of more employment opportunities (20).

Biodigested slurry can enhance the nutrient status of the soil by enriching the soil with all the major nutrients. This bioenergy production ensures the efficient use of renewable energy and promotes rapid decomposition of residues, leading to higher yield of organic fertilizer (21). Though biodigested slurry enhances the soil's physical, chemical and biological properties, it can also lead to accumulation of heavy metals in soil, in varying proportions depending on the source material. Thus, it is advisable to pretreat effluents and residues before digestion. The four important stages in biomethanation process are depicted in Fig. 1. This review provides insights into various physical, chemical and biological pretreatments available for the removal of these inhibitors, promoting the efficiency of the biogas production (Fig. 2) and supporting the sustainable concept of a circular economy.

Agricultural wastes

India produces about 352 million tons of agricultural waste, including straw, stover, cob and bagasse, which are often burnt, increasing the greenhouse emissions (carbon dioxide, nitrous oxide, nitrogen dioxide), causing respiratory diseases and environmental pollution. Instead of burning these resources, they can be efficiently bio-digested to produce biogas and bioethanol (22). The major factors in agricultural wastes that impede the biomethanation process are cellulose, lignin, C:N ratio and pH affecting the quality and quantity of

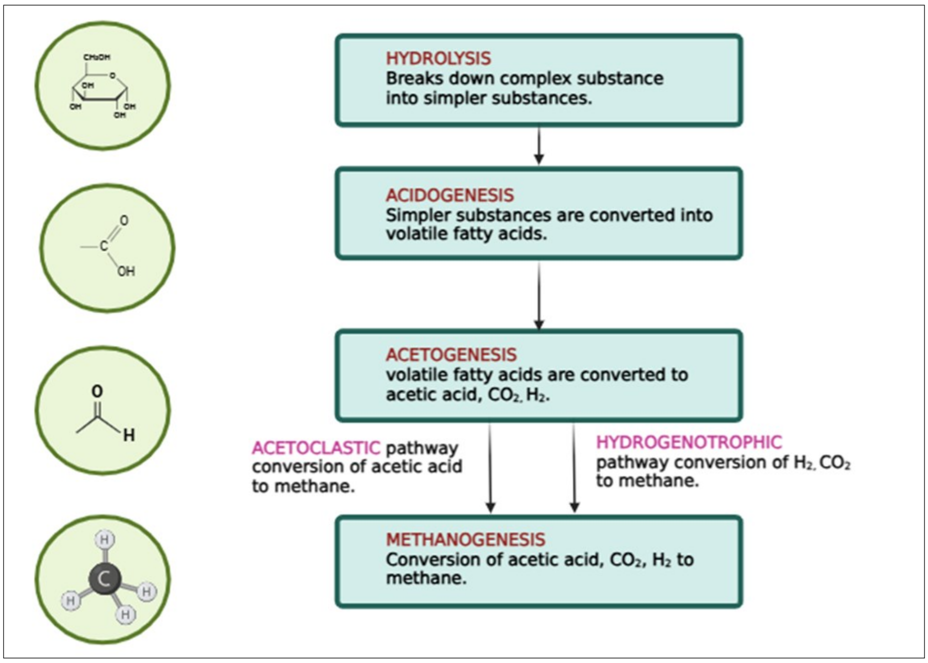

Fig. 1. Stages in biomethanation.

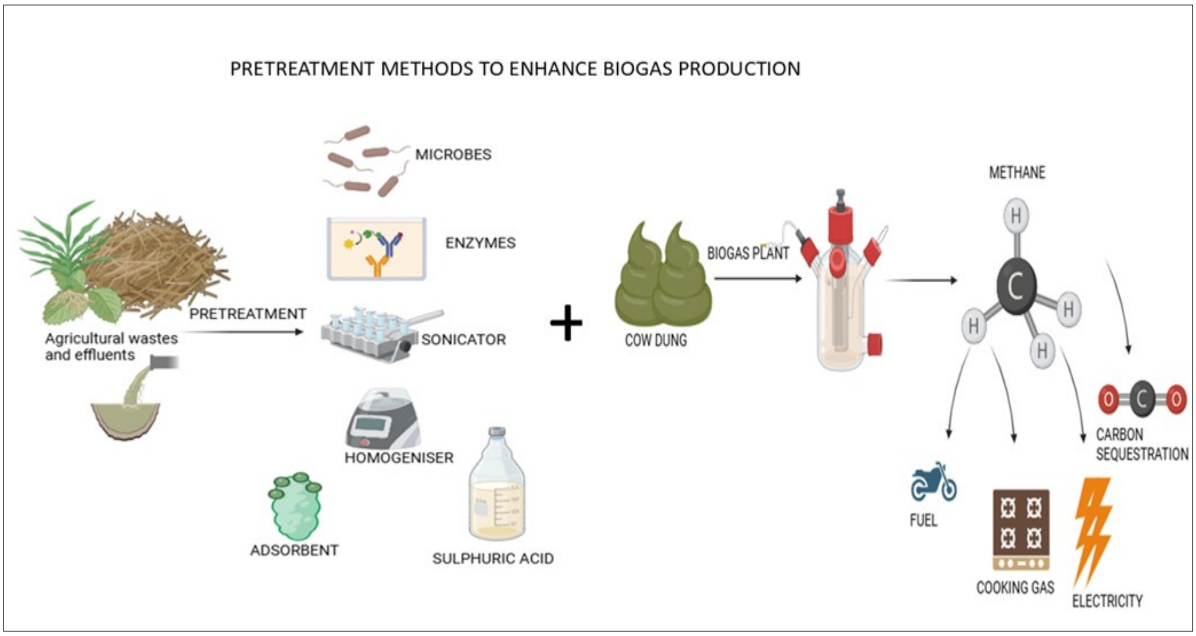

Fig. 2. Stages in biogas production.

biogas produced. Cellulose a polysaccharide abundantly found in agricultural residues, is degraded to produce anaerobic by-products such as bioethanol, methane, etc. Lignin is the hardest and the most impermeable layer in the plant cells, impeding the biomethanation process (Fig. 3) (23). Agricultural wastes are digested to produce methane, along with the formation of organic acids as intermediates. The major advantage of biogas production is recycling the wastes produced from animal husbandry, thereby ensuring the their proper utilization (24). Various pretreatment methods used to enhance biogas production from agricultural wastes are discussed in the section below. Table 1 provides various pretreatment methods for agricultural wastes.

Physical pretreatment

Mechanical pretreatment

Disk milling was observed to increase the fermentation of sugarcane (25). Mechanical pretreatments reduce particle size and increase surface area, thereby facilitating microbial degradation of lignocellulosic materials. Techniques include grinding, milling and shredding improve anaerobic digestion and increase biogas production from pretreated agricultural

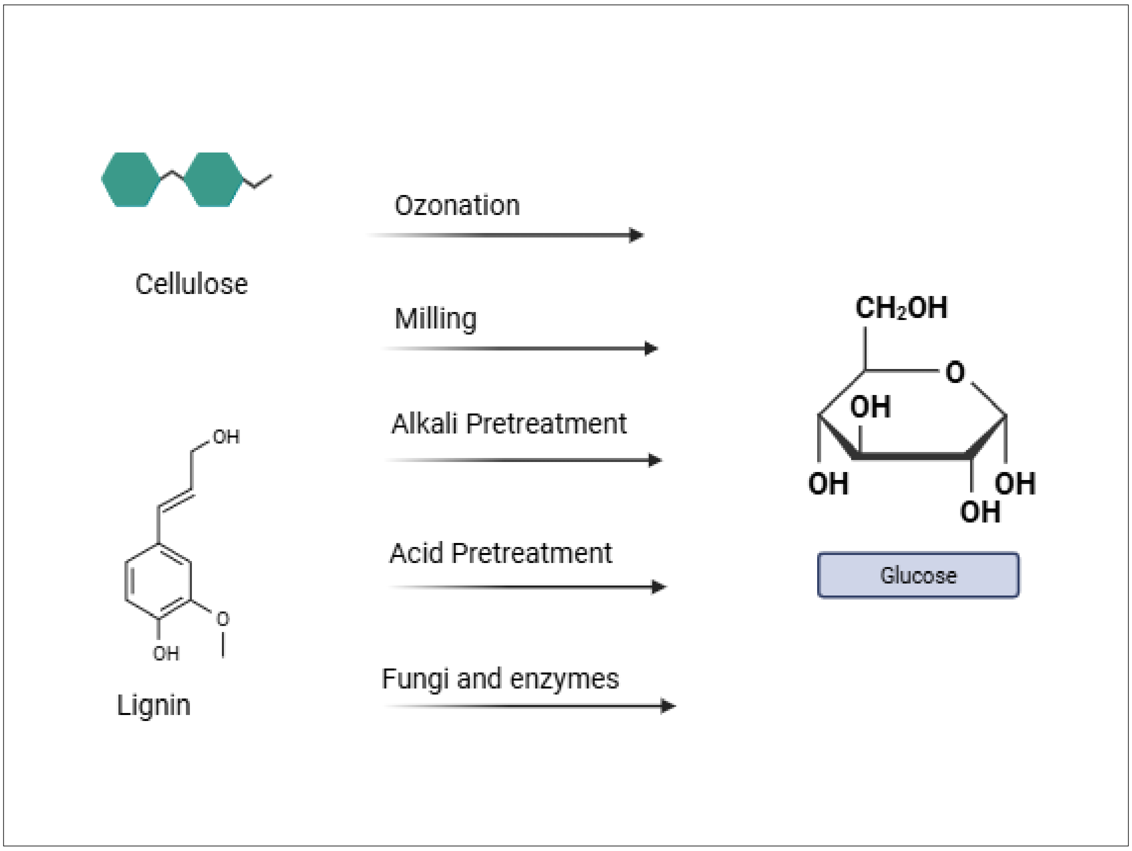

Fig. 3. Pretreatment of agricultural wastes.

Table 1. Various agricultural wastes, their inhibitors, efficiency of pretreatment methods and increase in biogas production

Agricultural wastes	Inhibitors	Method of removal	Efficiency	Increased methane Production	Pros and cons	References
Rice straw	Lignin, cellulose, hemicellulose	Pretreatment of paddy straw with microbial consortia for 48 hours. Thermally pretreated paddy straw at temperature of 80°- 120 °C for 1 hour used for anaerobic digestion.	Enhanced methane yields; degrades the polymeric structure of wastes. Partial degradation of wastes and reduces COD.	Consortium enhances upto 290 L N CH_4 kg^{-1} volatile solids. Enhances methane by 56 % (15 ml g^{-1} of volatile solids)	Time consuming method. Eco friendly technique. Efficient method.	(84, 85)
Wheat straw	Lignin, cellulose, Hemicellulose	6 % KOH was added to wheat straw enhanced anaerobic digestion.	Removal of 86 % of total solids, 89 % of volatile solids and 22 % of hemicellulose.	Enhances 45 % of biogas production. Improves biogas production	Simple and efficient method. Feasible method.	(86)
Maize	Lignocellulose	Steam pretreatment at 173 °C for 15 minutes at thermophilic condition.	Reduces lignin content.	Improves methane production by 637 LN m^{-3} day^{-1}	Less time required, efficient method.	(56)
Cotton	High cellulose content	Hot water along ultra-sonication used as pretreatment for cotton substrate to improve biomethanation. Hot water treatment combined with 6 % NaOH treatment along with addition of inoculum above the ratio of 0.4 (substrate: inoculum) enhances biomethanation.	Cellulosic substances are deploymerised. Biodegradability of cotton stock is improved.	Enhances biogas production 52.4 %. Biomethane production of about 111.8 ml g^{-1} of volatile solids.	The concept of circular economy is sustained. Inoculum addition is limiting step in the process.	(87,88)
Sugarcane	Lignin, cellulose, hemicellulose	Thermal pretreatment (180 °C) and NaOH of concentration 1.5M for the duration of 20 minutes enhanced anaerobic bioconversion of the sugarcane bagasse wastes.	Population of *Defluviitoga Methanothermobacter* are high in that treatment.	Production of high quantity fermentable sugars like glucose.	Low product concentration produced affects economic competitiveness.	(89)
Tofu wastes (soyabean)	Ammonia, high in organic content	Exposing tofu wastes to 140 °C for 15 mins improved methane production than control.	Batch process reduce acidification of ammonia and continuous process produces resistant methanotrophs.	Improves biogas production upto 589 ml g^{-1} of volatile solids.	Adaptable technique	(90)
Sunflower	Lignin, cellulose, hemicellulose	Mild thermal pretreatment at 55 °C for 24 hours followed by 4 g of sodium hydroxide treatment per 100g of total solids improved anaerobic digestion.	Breaks down lignocellulosic compounds present in stalk.	Improved methane production by 29-44 %	CSTR showed inhibitory effect on methane production by methanogens over the pretreated substrates.	(91)
Lucerne	-	Pretreated lucerne with 0.5 ml of alpha amylase enzyme improved anaerobic digestion of the wastes.	Improved anaerobic digestion.	Increases methane yield by 19.95 %	Low organic matter and high retention time are the major drawbacks.	(92)
Piggery waste	Ammonia	Micro aeration followed by retention in pre-hydraulic chamber for 1 day at 45 °C and in hydraulic chamber for 4 days at 45 °C improved the biogas production.	Removes ammonium, -	Increased methane by 20 %	Efficient method, time consuming.	(93)
Slaughter house wastes	BOD, COD, TSS, TN, TP	Stirring of waste water for 20 minutes at 26.43 °C, maintaining pH 7 and C:N ratio at 9.01 improves the anaerobic digestion of the wastes.	Reduced BOD 72 %	Improved bio-methane production by 170.26 L kg^{-1} of rumen.	Implicated in Indonesia could be applicable in other countries too.	(94)

wastes (26). Mechanical pretreatments such as knife milling, hammer milling and extrusion, have been shown to enhance methane production rates from 4 % to 48 %. This increase in methane production was attributed to the reduction in the size of wastes (27). The briquetting technique of straw has also gained momentum for increasing biomethane production while reducing transportation costs, storage losses and overall costs by 42.6 % (28).

Irradiation pretreatment

Gamma irradiation at 100 kGy combined with 2 % NaOH altered the cellulose, hemicellulose and lignin content, enhancing the enzymatic hydrolyses of agricultural wastes. Irradiation treatment using X-rays or gamma rays followed by chemical or enzymatic treatment, increased the degradation of agricultural wastes (29). Ionizing radiation such as gamma irradiation increased the disintegration of organic compounds. Treatment with 8.28-kGy radiation enhanced biogas yield by 14 %. Irradiation increased the enzymatic hydrolysis efficiency of rice straw, corn stalk and rapeseed straw at 800 kGy. This technique can thereby be used to enhance anaerobic digestion and improve biogas production (30).

Thermal pretreatment

Thermal pretreatment is an efficient method for the removal of lignin in agricultural wastes. The hard cell wall of sugarcane is disintegrated using hot water at a temperature of 200 to 220 °C, without affecting the sugar content (25). Thermal pretreatment is an effective method to improve the methanation of agricultural wastes. Studies have proven that heating agricultural residues at temperatures ranging from 90 to 200 °C enhances methane production (31). Freeze-thawing is another method, where agricultural wastes are exposed to a temperature of -20 °C, followed by potassium hydroxide treatment, increasing biomethane production (32).

Chemical pretreatment

Acid pretreatment

Free nitrous acid at different concentrations has been shown to effectively break down cellulose, thereby enhancing methane production. Free nitrous acid at a concentration of 2.31 mg improved methane production by up to 51 %. Acids like hydrochloric acid, sulphuric acid, phosphoric acid and nitric acid are used to depolymerize the cell walls of agricultural wastes. However, the diluted forms of acids is recommended to avoid corrosion, toxicity and inhibition of microbial growth (34). Free nitrous acid pretreatment of agricultural wastes increased biogas yield from wheat straw by about 32.24 % (9). Citric acid has also been used to degrade lignocellulosic compounds, thereby enhancing biomethane production (35).

Alkali pretreatment

The addition of 1 % sodium hydroxide and 1 % sulphuric acid to corn cobs has been shown to enhance microbial activity and improve anaerobic digestion (36). Cotton stalk pretreated with low concentrations of sodium hydroxide, followed by acid digestion and microbial inoculation, exhibit improved fermentation of the wastes. (37). This pretreatment is efficient in disintegrating cellulose, lignin and hemicellulose using sodium hydroxide, calcium hydroxide and sodium carbonate. Wheat straw is treated with sodium hydroxide and sodium carbonate, followed by alkaline hydrogen peroxide to enhance the anaerobic digestion (38). Agricultural wastes pretreated with ultrasound waves at 24 kHz, followed by sodium hydroxide, demonstrate enhanced lignin degradation, thereby supporting the improvement of the anaerobic digestion (39). Compared to acid treatment, alkali treatment is more efficient due to its lower toxicity and reduced environmental impact; however the process is time-consuming (40).

Oxidative pretreatment

Oxidative pretreatment involves oxidative properties of ozone, hydrogen peroxide and carbon dioxide, which cleaves the bonds present in cellulose, hemicellulose and lignin, thereby enhancing the degradation process. Hydrogen peroxide, along with peracetic acid and sodium hydroxide, is used to pretreat agricultural wastes, enhancing the efficiency of the lignin and hemicellulose degradation (38). Supercritical carbon dioxide has been used to digest green coconut fiber, disintegrating lignin and hemicellulose. This treatment enhances the anaerobic digestion of the green coconut fiber (41). It is an easy and eco-friendly method, but the cost is too high and removing the solvents used in the treatment remains a challenge (42).

Biological pretreatment

Fungal degradation

Fungal pretreatment is the most effective methods for degradation of hard agricultural wastes. White rot fungus, brown rot fungus and soft rot fungus have been used for degradation of lignin. Fungal treatment does not require any special instruments or machines. It is the most effective and efficient method. Factors affecting fungal treatment include moisture content, size of the wastes, reaction temperature and the incubation time (43). *Cerrena unicolor* is one of the most effective fungal strains used in the degradation of the lignin in rice straw through the production of a large quantity of laccase enzyme (44). Fungi like *Orpinomyces joyonii* has enhanced biogas and methane production from rice straw by 25 % and 38 % respectively (45).

Bacterial degradation

Bacterial strains like *Alcaligenes*, *Arthrobacter, Nocardia, Pseudomonas* and *Streptomyces* can easily degrade single-ring aromatic agricultural wastes using the lignin-degrading enzymes (46). The efficiency of the bacterial degradation of agricultural wastes ranges from 10 % to 89 %, with the majority of the bacteria falling under Proteobacteria, Firmicutes and Actinobacteria (47). Digestion of agricultural wastes along with shrimp chaff increased methane production up to 8.47 fold, wherein *Proteiniphilum* and *Methanosarcina* were the major contributors for anaerobic digestion (48). *Agrobacterium* sp. improved the anaerobic digestion of wood and newspaper waste, enhancing methane production by twofold (49). Ligninolytic bacteria such as *Bacillus* sp. has been isolated and added to paddy straw to improve biogas production (50).

Enzymes

Lignin-degrading enzymes like lignin catalases, peroxidases and laccases have been used directly for the degradation of wastes (51). Glycol oxidase, glyoxal oxidase and super mutase are the supportive enzymes that facilitates lignin breakdown indirectly, contributing to the value addition of agricultural wastes. Adding lignin peroxidase increased methane production by about 20 % in newspaper wastes, though it impeded the effect on softwood wastes (49). Enzymatic treatments enhanced digestion, increasing methane yield from 0.3 % to 21.1 %. Though enzymatic treatment improves biomethanation, factors like substrate type, enzyme mixture and dosage influence the effectiveness of anaerobic digestion of agricultural waste (52).

Physio-chemical pretreatments

Steam pretreatment

Rice straw was steamed at 200 °C for 10 mins, which helped degrade cellulose and hemicellulose, thereby enhancing the efficiency of anaerobic digestion (53). High-pressure steam can breakdown the lignocellulosic biomass, accelerating the degradation of agricultural wastes (54). Rice straw exposed to the temperature of 160 to 240°C for 12 - 30 mins improved biogas production by up to 32 % compared to un-treated straw (55). Wheat straw can be used as a better feedstock for biogas production when autoclaved at different retention times ranging from 30-60 mins. Autoclaving for 60 mins improved the biomethane production significantly (56).

Extrusion

Extrusion is the physio-thermal mechanism used for the degradation of agricultural wastes. It is a continuous and versatile process, with catalytic extrusion being the most preferred method (57). The extrusion process is an efficient process for recycling wastes into useful by-products, thereby reducing environmental pollution and supporting the concept of a circular economy (58). Twin-screw extrusion of agricultural wastes improved biomethane production by reducing the size and crystallinity of wastes (59). Pretreated corn straw moistened to 25 % and extruded at 110 rpm, improved methane and biogas production (51.63 %) compared to un-treated corn straw (49.57 %). The speed of the blade and the moisture content of the substrate affect the efficiency of the extrusion process (60). Table 2 provides a comparison of various pretreatment methods used for agricultural wastes.

Industrial effluents

Industries produce about 67000 million litres of effluents and sewage discharge mounts to approximately 23000 million litres per day. Of this sewage discharge, around 37 % comprises industrial effluents, which are discharged into the environment, leading to pollution (61). Paper mill effluents rich in potassium, chlorine, pH, color, total solids (TS), total dissolved solids (TDS), total suspended solids (TSS), biochemical oxygen demand (BOD) and chemical oxygen demand (COD), discharged into water bodies continue to increase pollution and bio-magnification (62). In the leather (tannery) industry, processing one metric ton of raw material into 20 % of finished product, generates more than 60 % of solid and liquid wastes, which can degrade the environment. About 50000 litres of wastewater rich in chromium (more than 5kg) are discharged into the water bodies, causing serious environmental pollution and various health issues to mankind. These effluents can be pretreated and converted into value-added products like biogas, bioethanol and other beneficial products (63). These industrial effluents are subjected to several physical, chemical and biological methods. These pretreated wastes are then anaerobically digested to produce bio-ethanol, biogas and nutrient-rich sludge for effective recycling (Fig. 4). This pretreatment reduces the pollution load and ensures the

Table 2. Comparison between various pretreatment methods used for agricultural wastes

Pretreatment methods	Efficiency	Advantages	Disadvantages	References
Mechanical method	Increases methane production within 4 % to 48 %	Increases the surface area by reducing the size and reduces the transportation charges	Requires labourers and machines and has a high chance of pollution and contamination, operation costs and time consumption is high	(26-28)
Thermal method	An increase in temperature increases the rate of degradation of lignin, cellulose followed by chemical pretreatment improving the efficiency of digestion	Removes the hard components of wastes, increases the degradation thereby increasing methane production	Losses of sugars and more quantity of water are required for the cooling process, at times the quality of produce is also lost	(25,32)
Acid degradation	Breaks down the rigid components of agricultural wastes improving the further degradation and methane production	The fastest method requires less time and space. Cost is comparatively less	Highly concentrated acids are corrosive and toxic to humans and the environment	(9,36)
Alkali Degradation	Disintegrates cellulose, hemicellulose and lignin from agricultural wastes	Less costly and is less harmful when compared to acid digestion	Time-consuming method and requires a combination of other physical methods	(39,40)
Steam exploitation	Similar to thermal pretreatment. Wet air has high penetration power and disintegration ability	The cheap method does not cause any pollution and the reusability of water is also possible	Applicable only for pressure and steam-tolerant wastes, require water and various equipment for operation	(54,55,56)
Extrusion	Reduces the size thereby increasing the surface area	Requires a combination of either physical or biological methods, efficient methods to scale down the size and improve the digestion	Costlier method, moisture content and speed of the blade (rpm) must be taken into consideration for different wastes	(57,60)
Bacteria	Enzymatically degrades the hard components of these wastes followed by digestion	The cheapest method causes less pollution to the environment	Pathogenicity of the organism involved, time and space constraints are the major issues here	(47,50)
Fungi	The production of enzymes degrades agricultural wastes	Cheapest method	Pathogenicity and spore production are the major issue	(43,45)

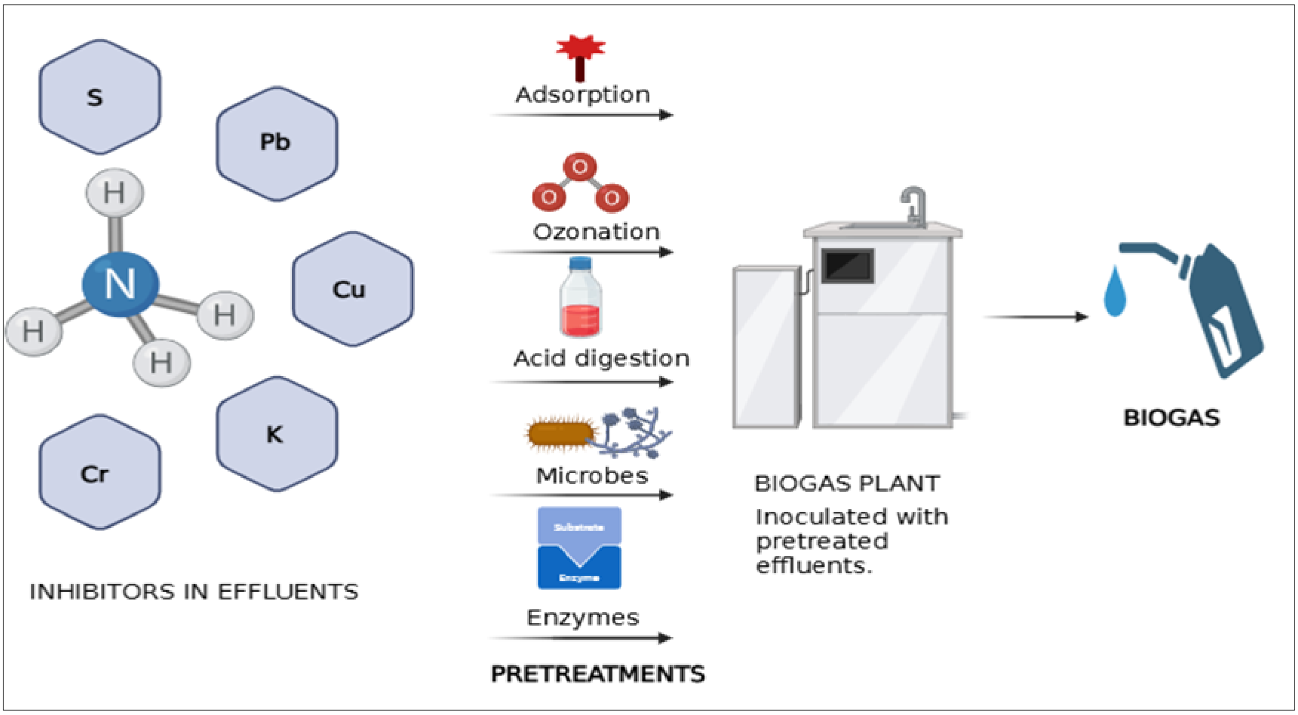

Fig. 4. Various pretreatment methods for industrial effluents.

Table 3. Various industrial effluents, their inhibitors, efficiency of pretreatment methods and increase in biogas production

Industries	Inhibitors	Pretreatment methods	Efficiency	Increased biogas production	References
Distillery	COD, phosphorous, potassium, BOD	3 phase pre-digestion using *Aspergillus oryzae, Neurospora* and anaerobes aided in production of mycoprotein and biogas.	COD was scaled down about 56.8 %, 11.8 % and 28.1 % in each phase of digestion.	Aided in production of mycoprotein (17.8 kg) and biogas (5 N m^3 cm^{-3} of waste water)	(95)
Vinasse	Sulphate	Ultrafiltration technique is used remove sulphate from the effluent followed by integration of anaerobes improves the biogas production.	Ultrafiltration has highest degradation rate and lag phase time for anaerobic digestion.	Increased the biogas yield by 4 %	(96)
Textile	COD, BOD sulphate	Pretreating the effluent with enzymes like alkaline peptidase improves the degradation of chemical substances improving the anaerobic degradation of wastes.	Removes the inhibitors effectively.	Increased the methane yield by 36 %	(97)
Tannery	Nitrogenic compounds, sulphur	Hybrid linear flow reactors are used along with specific microbial consortia for the effective generation of biogas.	Removes sulphur and other toxic nitrogenous compounds	Bio-methanation efficiency has been improved to 321 mL g^{-1} of COD consumed.	(98)

safe discharge of effluents. Table 3 provides a list of various industrial effluents and their pretreatment methods.

Pretreatment methods

Adsorption

Biosorption of heavy metals like Cd, Ni, Pb, etc. using brown algae and sugar beet pulp has proven effective in promoting microbial growth in industrial effluents, thereby facilitating their bioconversion into ethanol and methane (64). Hybrid adsorption or a combination of adsorption and ultrafiltration techniques improved the efficiency and effectiveness of the heavy metal removal. Additionally, the removed N-P-K were characterized and repurposed as soil improvers to enhance soil health and fertility (65). Adsorption is the process of accumulation of materials at the boundary between the two phases (solid and liquid). Any material that is found at the interface is known as the adsorbate and the solid on which adsorption takes place is known as adsorbent. Adsorption can be categorized into two types namely, chemisorption (irreversible due to ion exchange forming chemical bonds) and physisorption (reversible as it forms weaker van der Waals bonds). Activated carbon remains the most used adsorbent for industrial effluents (66). Vegetable wastes were dried and processed into activated carbon for the removal of COD, TSS, turbidity and oil or grease content. Ultrafiltration has also been employed to improve removal efficiency of heavy metals, potassium, etc. (67). Additionally, carbon nanotubes have shown significant capacity to adsorb heavy metals. Hydrogel adsorption is an emerging technology used for the removal of lead from industrial effluents. Treatment of hydrogel at 25 °C for a duration of 8 hrs has been reported to be effective in removing lead contaminants in wastewater (68).

Electrocoagulation

Coagulation with aluminum sulphate was reported to be the best pretreatment for the effective removal of inhibitors in the effluents. Pretreated industrial effluent was fed into an up-flow anaerobic sludge blanket (USAB), thereby improving the removal of pollutants in two steps - coagulation and ultra-filtration (69). Electrocoagulation (EC) improved biomethanation of industrial effluents by about 18 % and enhanced the removal of pollutants

(70). Electrocoagulation combined with microbial fuel cells improves the biogas production from various treated effluents (71).

Ozonation

Oxidation of organic matter in industrial effluent is the costliest method. To overcome this disadvantage, ozone with an oxidizing potential of 2.07 V reacted directly with molecular substances or through the generation of free OH radicals to break down the complex organic molecules into simpler ones in industrial effluent (72). Micro nanobubbles are used to prolong the reactivity of ozone to degrade organic compounds present in industrial effluents. This method is efficient in preventing groundwater contamination (73).

Chemical pretreatment

Chemical pretreatment is an effective method to remove various inhibitors present in industrial effluents. Pretreated effluents were subjected to anaerobically digestion to produce biogas or bioethanol. Recovered inhibitors like potassium and sulphate are used as enriched fertilizers by the farming community to reduce the cost of cultivation. The disadvantage of this method is that the use of chemicals at higher concentrations may lead to environmental pollution (74). The modified zeolite (zeolite impregnated with metal oxides of iron, aluminum and manganese) was treated with wastewater, which effectively recovered potassium and ammonium and was subsequently applied as fertilizers to improve soil health (75). Wastewater produced from the oil and gas industry is pretreated with sodium carbonate to precipitate calcium present as calcium carbonate. The efficiency of struvite precipitation increased at an optimum pH of 9.5, with an Mg: N: P ratio of 1.5:1:1.5, along with calcium pretreatment. Ammonium, potassium and magnesium were recovered at the rates of 85.9 %, 24.8 % and 96.8 % from struvite precipitation. Calcium pretreatment removed about 96 % of calcium and reduced magnesium loss by about 31.3 %. These recovered inhibitors are used as slow-releasing fertilizers (76). Sulphuric acid is used as a scrubbing agent in the stripping column for the efficient removal of ammonium from wastewater and enhance subsequent anaerobic digestion of the effluent (77).

Biological pretreatment

Microbial inoculants and enzymes

Bacterial strains *Bacillus* and *Escherichia coli* were used for the treatment of industrial dyes. The optimum conditions recorded for bioremediation were 28.3 °C for 1 day at pH 7.9 for *Bacillus* and 39.9 °C for 1 day at pH of 7.23 for *E. coli,* for the effective removal of dyes, thereby enhancing anaerobic stability of the methanogens (78). Seaweeds have high potential for applications in pharmaceuticals, cosmetics, food and bioremediation. The use of seaweeds as sorbents for the removal of copper from industrial effluents was efficient by up to 80 %. This pretreated effluent used for biogas production improved the efficiency by 5.2 %. Thus, integrating seaweeds in the pretreatment of industrial effluents is an emerging green technology for the removal of contaminants (79). Table 4 enlists various microbes and enzymes used in the pretreatment process. Table 3 provides a list of various industrial effluents and their pretreatment ways.

Future prospects and challenges

Usage of agricultural wastes as adsorbents for removal of the heavy metals and other toxic materials from industrial effluents is an effective strategy to improve further decomposition of the wastes. However, the disposal of spent adsorbents poses a problem to the environment. Therefore, by optimizing the techniques for effective disposal of both adsorbents and adsorbates, the biosorption stands out as the most efficient method for the removal of inhibitors in industrial effluents, thereby enhancing the production of biogas. Instead of burning agricultural wastes, they can be anaerobically digested with proper pretreatment for increased production of biogas. Further technologies could be developed for utilizing agricultural wastes as adsorbents to remove the inhibitors from the effluents, ensuring safe disposal and maximising their potential for the green energy production. Emphasis on circular economy must be strengthened for the sustainability of the environment and agriculture. The major challenge in the utilization of these wastes and effluents lies in the presence of inhibitors impeding the transformation process. These inhibitors can be effectively scrubbed using a combination of techniques to valorize waste to wealth.

Conclusion

Agricultural waste management has become a potent component of sustainable agriculture. Biomethanation of agricultural wastes and industrial effluents to produce renewable biogas technology has become imperative in the current situation. The over-exploitation of fossil fuels has resulted in the depletion of natural resources, amplifying the global energy crisis and escalating energy demand. Industrial wastewater discharged into the water bodies causes pollution and eutrophication. To reduce adverse effects, anaerobic digestion of these wastes has emerged as an effective solution. However, a few components present in the agricultural wastes impede the biomethanation process. The quantity of biogas generated is improved by pretreating these wastes physically, chemically or biologically. The method of pretreatment is chosen based on the waste type, to remove the inhibitors such as lignin, potassium, chlorine, sulphate, etc. These methods enhance the biomethanation process for agricultural wastes and industrial effluents, thereby increasing biogas production, scaling up the circular economy (Fig. 5) and promoting sustainable green agriculture towards a cleaner environment.

Table 4. Enzymes and their effect on degradation

Organisms	Enzymes	Effects	References
White rot Fungi	Oxidoreductases Manganese peroxidases	Biodegradation of organic pollutants. Reduces phenolic, non-phenolic lignocellulosic content of the wastes	(80) (81)
Azadirachta indica (neem) *Trametes hirsuta (*BM-2)	Catalases, peroxidases Super oxidase dismutase Laccase	Removes lead from municipal waste and industrial waste water. 65.5 % of phenol and 68.8 % color removal of industrial effluents.	(82) (83)

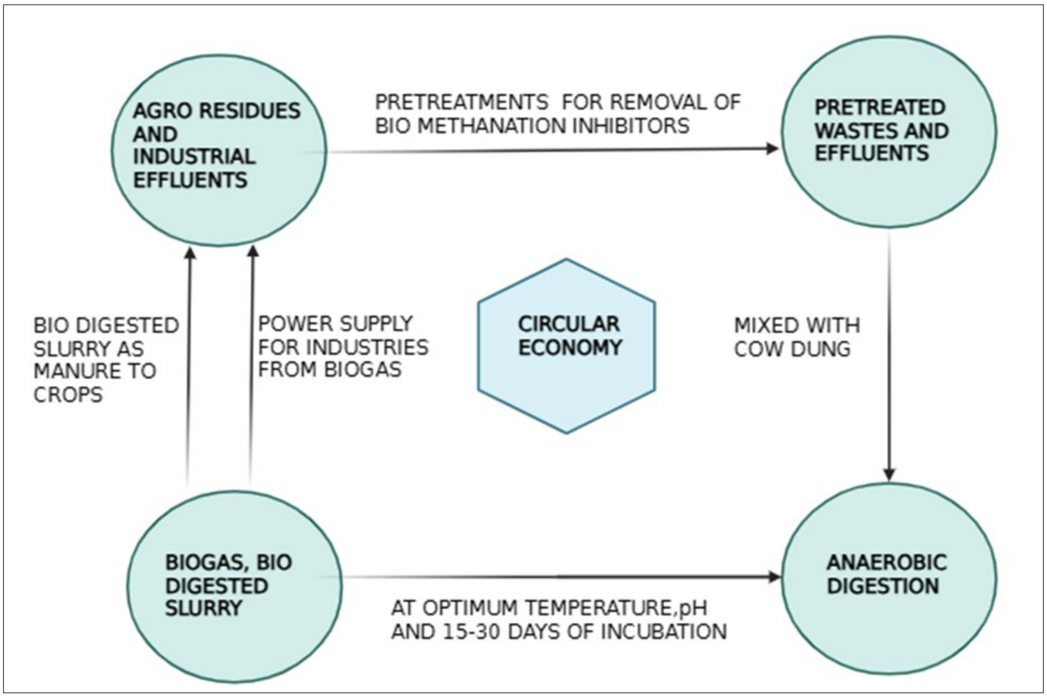

Fig. 5. Concept of circular economy.

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Authors' contributions

SV and RPD conceived the concept and wrote the manuscript. SV designed the tables and diagrams. SK[1], SK[2], DR and AB reviewed, revised and finalised the manuscript. All authors read and approved the final manuscript [SK[1] - Senthilraja K; SK[2]-Subburamu Karthikeyan].

Compliance with ethical standards

Conflict of interest: The authors declare no conflict of interest.

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