sustainability

Review

On the Conversion of Paper Waste and Rejects into High-Value Materials and Energy

Hatem Abushammala [1,*], Muhammad Adil Masood [1], Salma Taqi Ghulam [1] and Jia Mao [2]

1 Environmental Health and Safety Program, College of Health Sciences, Abu Dhabi University, Abu Dhabi 59911, United Arab Emirates
2 Department of Mechanical Engineering, Al Ghurair University, International Academic City, Dubai 37374, United Arab Emirates
* Correspondence: hatem.abushammala@adu.ac.ae; Tel.: +971-0-25015290

Abstract: The pulp and paper industry (PPI) is a major contributor to the global economy, but it also poses a challenge for waste disposal, as it generates large amounts of several waste streams. Among these, paper rejects are generated during the papermaking process and could account for up to 25% of the produced paper. Moreover, hundreds of millions of tons of paper are produced annually that end up in landfills if not burnt or recycled. Furthermore, the PPI significantly contributes to climate change and global warming in the form of deforestation and water and air pollution. Therefore, the impact of this industry on the sustainability of natural resources and its adverse environmental health effects requires special attention. This review focuses on discussing the sustainable routes to utilize paper waste and rejects from the PPI towards a circular economy. At first, it discusses the industry itself and its environmental impact, followed by the possible sustainable approaches that can be implemented to improve papermaking processes as well as waste management systems, including paper recycling. The literature indicates that paper recycling is crucial because, if appropriately designed, it significantly lowers greenhouse gas emissions, water and resources consumption, and manufacturing costs. However, several concerns have surfaced about the different chemicals that are used to improve recycling efficiency and recycled paper quality. Furthermore, paper recycling is limited to up to seven times. This review, therefore, goes on to highlight several sustainable waste management routes for paper waste utilization other than recycling by emphasizing the concept of converting paper waste and rejects into energy and high-value materials, including biofuels, biohydrogen, biomethane, heat, nanocellulose, hydrochar, construction materials, and soil amendments. Both the benefits and shortcomings of these waste management routes and their applications are discussed. It becomes clear from this review that sustainable management solutions for paper waste and rejects are implementable, but further research and development are still needed.

Keywords: paper waste; paper reject; pulp; waste management; recycling; circular economy

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1. Introduction

Paper is produced from the cellulose pulp fibers obtained from wood pulping. It was first produced in China, spread to Europe, and has since been manufactured all over the world [1]. Over the years, paper manufacturing processes have changed in terms of the techniques and chemicals used, but wood remains the most widely used source of fibers in the industry [2]. Despite digitization, paper is still consumed in offices, magazines, and newspapers. It also contributes to 31% of the packaging market globally [3]. The pulp and paper industry (PPI) is a major contributor to the global economy despite the low-profit margins due to well-known challenges in pricing and fulfilling consumer requirements. This industry also faces constant pressure to reduce emissions that cause water and air pollution [4]. Deforestation is also a challenge for the PPI, as roughly 4 billion trees are cut down every year to serve as the raw material for paper making [5]. Most of the recent strategies for reducing this effect aim to increase the efficiency of printing techniques and the utilization of recycled paper [6]. The PPI is considered a major water polluter because it releases massive amounts of toxic substances into water bodies. The wastewater from the PPI contains more than 250 toxic chemicals resulting from various processes in wood pulping and papermaking, including organic, sulfur, and chlorinated compounds and heavy metals, such as chlorophenols, sulfides, iron, zinc, and manganese [7,8]. The PPI also generates various types of waste, such as wood rejects, black liquor, paper rejects, and sludge [9]. The large amounts of waste present a challenge for waste disposal and can result in significant damage to water, air, and soil. Furthermore, the organic matter in wastewater sludge, if discharged into water bodies, increases biochemical oxygen demand (BOD), causing eutrophication [10]. Waste combustion can also cause health problems in nearby communities. Recently, advanced incineration technologies have been developed and used, which emit less harmful gases, but the health effects of incinerators can still be difficult to evaluate because of the simultaneous pollution from many other sources, such as automobiles and industries [11]. The PPI is also energy intensive, which can be improved by implementing energy conservation technologies, using renewable energy, and utilizing the biomass rejects of the industry as bioenergy [9].

The increased awareness of environmental issues and the rapid depletion of natural resources has promoted the interest of industries to reuse the waste generated in their manufacturing processes [12–16]. In addition, strict governmental regulations have been enacted to facilitate the utilization of wastes (reuse, recycling, or conversion), which were formerly burnt or disposed of in landfills, to reduce their environmental impact on air, soil, and water resources [17]. Sustainable waste management is a basic part of sustainable development, as multiple benefits in the public health, safety, and environmental sectors can be gained. Sustainable waste management reduces greenhouse gas emissions, improves the quality of life, preserves natural resources, and reduces soil and water contamination [18]. Moreover, modern waste management has a major goal of effectively recovering energy from waste before safely disposing of it. For example, organic waste has a high energy recovery potential [19], and municipal solid waste is listed as a renewable energy resource by the United States Environmental Protection Agency [20]. The recovery aspect of sustainable waste management is based on a hierarchy of waste prevention, reuse, recycling, recovery, and disposal. It is therefore important to mention that, before incineration and disposal, no efforts should be spared to convert paper waste and rejects into materials that can function as starting materials or finished products for all possible kinds of applications [21].

Several reviews are available in the literature about paper waste. Some discussed specific paper waste management techniques, including sorting and recycling [22,23], while others focused on the conversion of paper waste into a specific product, including construction materials and nanocellulose [24,25]. Our review comprehensively discusses the possible routes for the efficient conversion of paper waste and rejects into both high-value materials and energy. First of all, the review starts by explaining paper production from wood, including wood pulping and papermaking, and its impact on the environment while describing possible emissions, effluents, and wastes. It then continues to describe the process of recycling paper waste and rejects and the environmental and economic advantages. It then discusses the possible routes to convert paper waste into energy and high-value materials when it is not possible to further recycle paper into high-quality paper products. Finally, it economically and environmentally compares these routes with a focus on composting, anaerobic digestion, and incineration. Overall, this review provides a comprehensive overview of paper's lifecycle—from wood to paper to energy and other useful products—towards a circular economy.

2. Wood Pulping and Papermaking

The PPI requires four main inputs for papermaking: cellulose fibers, chemicals, energy, and water. In the past, the industry utilized cotton and linen rags as cellulose sources but later shifted to wood for increased paper production and quality [26]. Cellulose is the most abundant biopolymer, and it is used for paper, cardboard, and textile manufacturing [27]. It is a linear homopolymer of glucose and represents around 40–60% of wood mass. The second most abundant biopolymers on Earth after cellulose are lignin and hemicellulose, each representing around 20–30% of wood mass [28]. Lignin is a networked polymer of phenylpropane units, and it is responsible for the resistance and stiffness of wood in living trees [29]. It also gives wood its yellowish to brownish color because it is brown in color when extracted as powder, while cellulose and hemicellulose are white to off-white [28]. Hemicellulose is a branched heteropolymer of seven simple sugars: glucose, galactose, mannose, xylose, arabinose, glucuronic acid, and galacturonic acid [30]. It acts as the glue between cellulose and lignin in the structure of wood at the anatomic level [31].

The production of paper from wood involves several steps: wood pulping, pulp bleaching, purification, beating and refining, papermaking, and final treatments [3]. Being the first step in papermaking, the main goal of the pulping process is to remove lignin and hemicellulose while keeping the structure of the cellulose fibers intact to form pulp [32]. The efficiency of pulp production is dependent on the chemical composition of the wood and the manufacturing processes used for pulping. The variation in the lignin content of wood significantly affects pulping because lignin is the most recalcitrant wood component to remove [33]. Wood pulping methods can be chemical, mechanical, or semi-mechanical [34]. As the names indicate, chemical pulping uses chemicals to dissolve lignin and hemicellulose to produce the pulp fibers; mechanical pulping uses mechanical processes such as grinding and refining for that purpose [35]. The pulp produced by chemical methods is of a higher quality and contains a higher amount of cellulose fibers compared to the mechanically produced pulp. In other words, chemical pulping is more efficient in removing lignin and hemicellulose, leading to a pulp with higher cellulose content [36]. Chemical and mechanical pulps are advantageous for different applications. For example, mechanical pulp has excellent printing properties [37]. Along with papermaking, chemical pulp is used in other applications, such as the textile industry, pharmaceutical industry, and fuel production [38]. Kraft pulping and sulfite pulping are the most common chemical pulping methods, with Kraft pulping being the most commonly used [39]. In 2016, 95% of the paper mills in North America used Kraft pulping [40]. Environmentally speaking, numerous waste products, such as lignin and effluent sludge, are generated during Kraft pulping [41]. After wood pulping, bleaching takes place to further eliminate the remaining lignin [40]. It improves the whiteness of mechanical or chemical pulps by removing the chromophoric lignin groups using chlorine or chlorine dioxide [42]. The bleached pulp is then purified by removing any rejected cellulose fibers via filtration. Rejected fibers exhibit similar properties to bleached pulp but do not hold a strong place in the global market due to the presence of impurities. Still, they are sometimes used as raw materials for cardboard manufacturing [41]. The purified bleached pulp then goes through a beating and refining process to increase the surface area of its fibers, which in turn increases their ability to hold water and form better bonds before the papermaking process [35]. The papermaking process is mostly a pulp dewatering process, in which the pulp is passed through rollers or wire meshes, which assist in the removal of water and the formation of paper. Finally, the newly formed paper is post-treated by calendaring, supercalendering, sizing, laminating, impregnating, or saturating to smoothen the surface of the paper and increase its quality [3,43].

The PPI has a significant impact on the environment as the main pulp fibers required for paper come from trees. Millions of trees are cut down annually to suffice the material uptake of the industry [44], and it is known that deforestation leads to an increase in temperatures globally, a rise in sea level, and a loss of biodiversity [6]. Moreover, pulping and papermaking require massive amounts of water, and the discharged wastewater contains high levels of toxic chemicals that harm the environment [45]. For instance, bleaching requires the use of different oxidizing agents, such as chlorine dioxide and ozone, which are considered harmful to the environment. In order to mitigate their environmental harm, the pulp is sometimes bleached using other green agents such as hydrogen peroxide;

the resulting pulp is then a chlorine-free pulp [40,46]. Green solvents such as ionic liquids have also been explored in the literature as pulping agents to replace the harsh chemicals used in wood pulping [47–51]. Wastewater from the PPI is treated either by aerobic or anaerobic biological treatment, with aerobic being the most commonly used to reduce its BOD [52]. Aerobic treatment requires oxygen, which is supplied by aeration equipment. Anaerobic treatment is carried out in the absence of oxygen and has several advantages over aerobic treatment, including lower sludge production, but it is not commonly used in the PPI due to the formation of hydrogen sulfide [53]. The most common methods of disposal of the final paper sludge are landfilling and incineration [54]. When paper sludge is incinerated for energy recovery, another waste stream of the PPI, known as paper sludge ash, is produced. While paper sludge remains the main waste stream of the paper industry, paper sludge ash is also produced in large amounts [54]. Paper mill sludges are a mixture of several substances, including inorganic solids, chemical additives, and paper fibers [55]. From an environmental viewpoint, incineration and landfilling of sludge have a negative impact on the environment due to air and water pollution and land usage [56]. To reduce its environmental impact, the PPI explored paper waste as a raw material to produce paper, which seems to be environmentally beneficial [22]. However, many concerns have surfaced about the chemicals that are added to recycled paper to improve process efficiency and product quality [57]. Furthermore, recycled paper might have more harmful chemicals present in it due to cross-contamination with other wastes [23]. Moreover, waste recycling does not form a complete cycle as the amount of collected paper waste does not meet the required demands of paper products considering that recycling is not 100% efficient, as some of the paper waste degrades during the process [23]. Recycling also does not significantly reduce the greenhouse gas emissions of the PPI because it is not usually properly performed [58] and is still an energy-intensive process [59]. Due to the increase in global recycling rates, the amount of paper sludge from deinking and repulping also increased [60]. Deinking sludge contains ink particles, short cellulosic fibers, coatings, and deinking additives. Most deinking sludge is dewatered and incinerated for volume reduction and energy recovery, and very small amounts of it are used for the manufacturing of cement and bricks [61]. Figure 1 shows a summary of the main waste streams that are generated during paper's lifecycle, starting with wood pulping to paper making, consumption, and recycling.

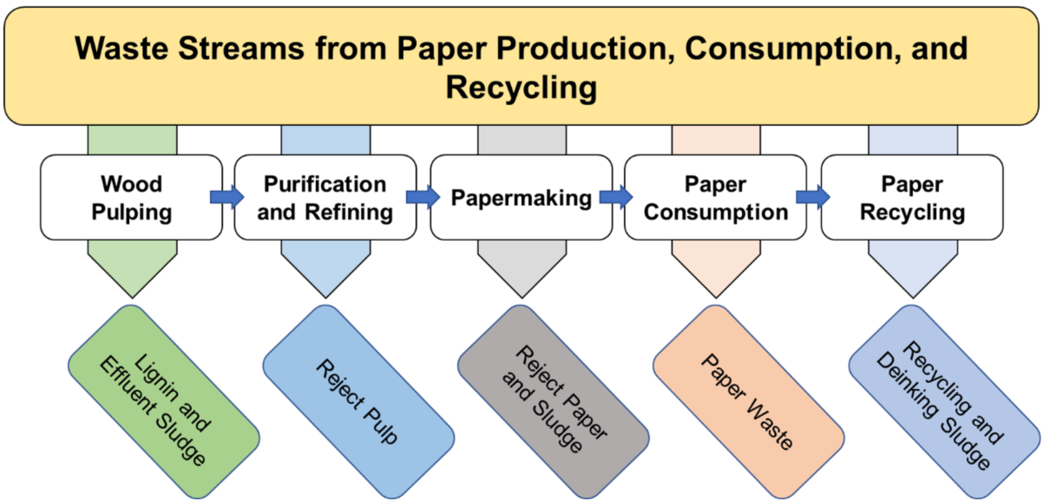

Figure 1. Possible waste streams generated during the lifecycle of paper from wood pulping to paper production, consumption, and recycling.

Overall, the PPI is a main consumer of wood, electricity, and water. It has a current objective of producing affordable and high-quality pulp while preserving natural resources [62]. Several parts of the world cannot keep up with the wood demand of the PPI and have shifted to alternative options such as bamboo, wheat straw, and rice straw [63]. The production of paper from non-wood sources has several advantages, such as ease of pulping and the high quality of the fibers, which can be used for the production of special types of paper [64]. Rice straw has a high cellulose content making it an excellent resource for paper production. Rice straw combined with wastepaper can produce paper that can be used as wrapping paper, art paper, and writing paper [65]. Moreover, non-wood fibers obtained from agricultural residues can solve the problem of agricultural waste disposal as it is commonly incinerated. Non-wood fibers also vary in length, which can be used to produce several types of paper [66]. Tea waste from the incorrect harvesting of black tea leaves, which is disposed of by landfilling, can also be used for papermaking [67]. Pineapple leaves and banana plant fibers are also good alternative raw materials for paper production as they both have high cellulose content [68].

3. Paper Waste and Rejects and Their Recycling

Paper can be virgin paper, which is produced from virgin pulp, or recycled paper, produced from recycled wastepaper or reprocessed pulp [3]. Paper recycling reduces the burden on the environment by minimizing natural resource consumption and reducing the amount of toxic chemicals released into the environment [59]. Paper recycling also plays a major role in decreasing greenhouse gas emissions since fewer trees are cut down, leading to more CO_2 captured through photosynthesis; paper manufacturing costs are also reduced because fewer processes are required [69,70]. A study conducted using material flow analysis and life cycle assessment on waste paper recycling in Spain showed that each ton of virgin paper emits around 530 kg CO_{2-eq} while a ton of recycled paper emits only around 210 kg CO_{2-eq}, reducing CO_2 emission by 320 kg CO_{2-eq} per ton of paper [71]. Recycling reduces pollution in water bodies by 35% and air pollution by 74%. Moreover, it reduces land space usage and conserves natural resources [72]. The main sources of paper waste for recycled papermaking are industries, households, and small businesses. Globally, approximately half of the paper waste is recycled, whereas half is incinerated or landfilled [73]. Households generally are a good source of mixed and graphic papers, whereas trade and industries provide corrugated paper used for packaging. Offices, along with printing and conversion operations, are a source of high-grade paper [74,75]. There are around 50 identified grades of wastepaper, and the quality of wastepaper is a determinant of the end quality of the recycled paper. However, purity and quality are negatively affected if different types of wastepaper are mixed together [72]. Wastepaper often contains substances introduced in the collection and handling processes known as non-paper components, such as laminated covers, staples, cosmetic samples, and plastic wrapping. These components can be removed in limited amounts during recycling, but cost increases if they are present in high amounts. This highlights the importance of an efficient waste collection and handling stage [74].

The paper recycling process involves several steps, including repulping, deinking, and refining. Repulping involves separating the fibers of the wastepaper using water and chemicals. Deinking and refining are not absolutely necessary steps for paper recycling, but they greatly increase the strength and appearance of the final product [74]. Deinking improves the visual quality of the recycled paper but does not whiten unbleached fibers. Therefore, bleaching may be needed. Several methods of deinking are used, with the floatation method being the most common [74]. The quality of deinked pulp is determined by several factors, including the printing method, composition of the ink, and recycling method. Paper printed using xerographic and inkjet technology with water- and toner-based inks further increase the issues in recycling systems. Paper printed by flexographic and offset methods should not be treated together as both require different deinking mediums [76]. Finally, the deinked pulp is refined to prepare it for the papermaking process [77]. Cellulosic fibers undergo different changes when recycled. After several recycling processes, they have a significantly low bonding potential compared to virgin fibers. This loss of bonding can be mostly restored by refining, but eventually, the fiber length decreases, affecting the strength of the paper [78]. It is important to mention that the effects of recycling on mechanical and chemical pulp are different. The bonding potential of chemical pulp significantly reduces due to the repeated drying and rewetting processes during recycling, while mechanical pulp deteriorates less and demonstrates a slight improvement in bonding potential [59]. For example, a study showed that the tensile index of sheets prepared from chemical pulp decreased from 65 to 55 N.m/g after one cycle of recycling, while the tensile index of sheets prepared from mechanical pulp increased from 30 to 35 N.m/g after one cycle of recycling [79]. In terms of the available technologies for recycling, one of the leading technologies is in situ precipitated calcium carbonate (PCC), which utilizes the CO_2 obtained from other industries with natural limestone to produce the calcium carbonate needed for paper production. This technology increases the strength and opacity of paper, reduces wastewater discharge, and saves energy, all of which result in lower manufacturing costs and minimal harm to the environment [73]. Another technology is dry paper recycling, which is used to recycle paper in offices without the need to export the wastepaper to a recycling facility. Dry paper recycling involves three main steps: separating the paper fibers by applying impact force, producing a sheet of paper with the collected fibers, and binding the fibers together using powdered binders and heat [80]. This technology reduces CO_2 emissions by 22% and water consumption by 99%. However, power consumption and environmental burden are significantly high considering the use of cartridges containing bonding agents. Therefore, further improvements are required to enhance its greenness [81].

Recycled paper is used for various applications such as newspaper, envelopes, office and printing paper, cardboard, and insulation. The utilization of recycled fibers depends on the quality requirement of the end product [82–84]. Paper production cannot fully depend on recycled paper since fresh fibers are required to add strength to newly produced paper and improve paper quality since some wastepaper grades cannot be used to produce high-quality paper. For instance, newspapers are produced using a high percentage (80%) of recovered fibers and 20% of primary fibers, while printing paper and magazine paper use 100% primary fibers, which are chemically treated due to required whiteness and brightness [4]. Moreover, the availability of waste paper is an issue since some types of paper, such as sanitary paper and cigarette paper, disintegrate with use, which makes them unsuitable for recycling [85]. Furthermore, paper can be recycled up to seven times, after which the cellulose fibers are deemed unsuitable for paper production and are rejected [4]. Therefore, there is a strong need for routes other than recycling for the utilization of paper waste, which is discussed below.

4. Conversion of Paper Waste into Energy and High-Value Materials

One major approach towards sustainability is the conversion of waste into energy and value-added products, which has shown significant development recently. While minimizing linear economies, a sustainable circular economy should encourage the steps of take, make, use, reuse, and recycle, as opposed to a linear economy that follows take, make, use, dispose, and pollute [86]. Below are examples of how paper waste and rejects can be converted into energy and useful products, including biofuels, biohydrogen, biomethane, heat, nanocellulose, hydrochar, construction materials, and soil amendments.

4.1. Biofuels

Bioethanol is the only fuel so far that can be transported in liquid form and does not release greenhouse gases into the atmosphere when burned as the CO_2 emitted is used up by plants in photosynthesis as long as the biofuel consumption rate is not more than the photosynthesis rate [87]. First-generation bioethanol is produced from sugar and starch, whereas second-generation bioethanol is produced from waste containing lignocellulose. Bioethanol production requires high amounts of cellulose, making paper waste a potential raw material [88]. The commercial production of bioethanol currently involves only first-generation product, which is produced by food crops such as sugarcane and corn as a feedstock. A major drawback of first-generation bioethanol is the usage of food crops for feedstock, which results in less available food and an increase in food prices. Bioethanol is currently the most produced biofuel globally, with the United States producing the majority of it [89]. To convert paper waste into bioethanol, cellulose and hemicellulose are hydrolyzed to simple sugars, either chemically or using enzymes. Enzymatic hydrolysis is used more commonly as it is more specific and milder [90]. The simple sugars are then converted into bioethanol by fermentation [91]. Most of the processes that are used currently for bioethanol production from biomass involve certain pretreatments before hydrolysis and fermentation. Pretreatments include the usage of acid, alkali, or other organic solvents, and choosing the correct pretreatment is crucial in bioethanol production as it affects the cellulose conversion rates and enzyme activity [87]. Pretreatment is essential to gain good yields of sugars from cellulose and hemicellulose, and it increases the digestibility of cellulosic biomass [92]. The chosen pretreatment should not degrade the sugars into enzyme activity inhibitors, use minimal energy, and require fewer chemicals. For example, pretreated paper sludge yields 90% or more sugar, whereas untreated sludge yields 20% or less [93]. The presence of barrier components such as lignin and the crystalline regions of cellulose decreases the effectivity of enzymes [92]. Newspaper and cardboard contain a high amount of barrier components, whereas hygiene paper and office paper have minimal amounts of them [94]. Using paper waste as a raw material for bioethanol production provides an alternate energy source and reduces the usage of fossil fuels, solving environmental issues such as global warming. It also solves issues related to biofuel production from biomass, such as land use, deforestation, threats to global food security, and the limited amount of available biomass [95]. It is, however, not as effective in fighting global warming as renewable energy sources such as solar and wind as it still emits greenhouse gases [96].

Paper waste has an advantage over other lignocellulose materials in bioethanol production as it does not require additional pretreatments since the majority of lignin is already removed in various paper manufacturing processes [97]. Moreover, the degradable nature of paper waste makes it suitable for bioethanol production. In addition to that, wastepaper is easily available at low costs and contains high levels of carbohydrates. Bioethanol production from wastepaper also provides an alternative to recycling since recycling has several limitations, such as additional energy usage in recycling processes and a limited number of recycling cycles [98]. The paper sludge from the papermaking process contains short-length fibers and has high amounts of lignocellulose and carbohydrates, which can also be used to produce bioethanol rather than incinerating or disposing of it. However, the presence of ash and calcium carbonate block the bioconversion of fibers as they increase the pH value of sludge, making it unsuitable for enzyme activity [99]. Treatments for calcium carbonate removal involve the usage of acids and produce CO_2, causing environmental issues. To overcome this issue, simultaneous saccharification and fermentation (SSF) can be used to neutralize the calcium carbonate present in sludge without needing pretreatments [100].

4.2. Biohydrogen and Biogas

The biohydrogen produced from biomass waste is a renewable source of energy and does not have a negative environmental impact. It has the potential to overcome several environmental issues and the global energy demand [101]. A major barrier in biohydrogen production is the cost, as it has complications in terms of storage, compression, distribution networks, and lack of durable fuel technologies [102]. Hydrogen fuel cells are also costly to set up, delicate, and do not function for extended periods of time [103]. Thermochemical pulping (TMP) is a papermaking process by which wood fibers are treated using hot steam under pressure. TMP results in anaerobically-treated wastewater to produce biohydrogen in a process called dark fermentation (Figure 2) [85]. Dark fermentation is a temperature-sensitive process since a slight shift in temperature can significantly increase or decrease the hydrogen yield; hence, the temperature of wastewater has to be controlled efficiently [104]. A group of researchers used recirculated two-phased anaerobic digestion to produce biohydrogen and biomethane simultaneously from municipal solid waste, 50% of which was paper waste [105]. Anaerobic digestion of paper is one of the major waste management options since it is not economically burdening and offers environmental benefits, producing renewable energy in the form of biomethane. Food waste and paper waste can be digested anaerobically individually (mono-anaerobic digestion) or combined (co-digestion). Co-digestion provides a better yield of methane compared to mono-digestion. Thermophilic (50–60 °C) and mesophilic (30–40 °C) are the two temperature ranges of anaerobic digestion, with thermophilic being better due to high methane gas content, low hydrogen sulfide content, and high rate of organic matter degradation [106]. When waste is anaerobically digested, methane is obtained in a high percentage (50–70%), followed by carbon dioxide and other gases such as hydrogen. Cellulosic wastes generally do not produce high yields of biogases due to their poor biodegradability. To overcome this, paper waste can be co-digested with other organic wastes to increase gas flammability [107]. A study conducted by Priadi et al. (2014) showed that the anaerobic digestion of paper sludge seeded with cow manure produced a significantly high yield of methane (269 mL/g) compared to the anaerobic digestion of paper sludge alone (14.7 mL/g). The treatment of wastewater originating from the paper industry results in large quantities of paper sludge containing chlorinated organics, pathogens, and traces of heavy metals [108]. Wastewater from industrial and municipal sources can be treated by several different anaerobic reactors, such as an up-flow anaerobic filter, modified anaerobic baffled reactor (MABR), or up-flow anaerobic sludge blanket (UASB). UASB is the most commonly used anaerobic reactor for municipal and industrial wastewater treatment since it is stable and consumes energy efficiently. Using this reactor, wastewater containing organic matter released from papermaking or recycling industries can be used to produce biomethane [109].

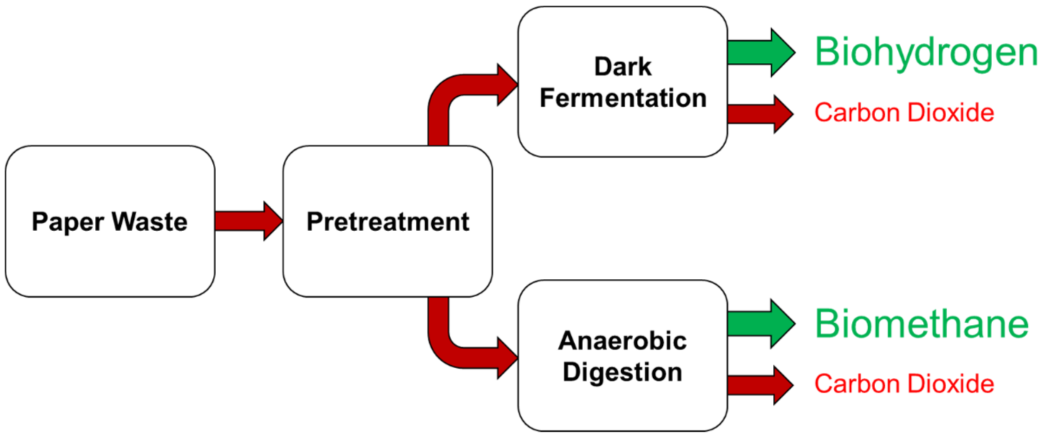

Figure 2. Conversion of paper waste into biohydrogen or biomethane using dark fermentation and anaerobic digestion, respectively.

Paper waste often undergoes pretreatments before anaerobic digestion to obtain a higher yield of biomethane [110]. Mechanical pretreatments such as shredding have shown no increase in biomethane production. Biological pretreatment, on the other hand, which consists of treating different types of paper waste with a thermophilic cellulose-degrading consortium, increases biomethane yield [111]. Moreover, biogas production is dependent on several different factors, such as the volatile solid content of the feedstock and the level of biological activity in the digester. Carbon and nitrogen balance in feeding material also affects biogas production, with the optimal carbon and nitrogen balance ratio being between 25:1 and 30:1. In addition, environmental factors, including temperature and pH, determine the obtained biogas yield [112]. Furthermore, variations in the pulp production procedures can alter methane production in anaerobic digesters. Primary pulp obtained from Kraft and sulfite pulping provides a high yield of methane when anaerobically digested due to the efficient removal of lignin from the pulp in papermaking processes. Another advantage of using these types of pulps is that methane production remains stable despite the shift in raw material or the pulp being in the bleached or unbleached form [113].

4.3. Heat by Incineration

Paper waste has three main disposal methods, landfilling, incineration, and composting. Landfilling is the most common disposal method among the three. The effectiveness of landfilling is severely reduced due to the extensive land usage, especially in countries such as China, which is one of the largest producers of solid waste globally [114]. Landfilling emits CH_4 as solid waste decomposes anaerobically. Paper waste can be utilized as renewable energy for power generation and minimizes the greenhouse gas emissions released from landfilling [115]. In the past, landfilling sites were available in close proximity to urban areas, which limited the development and need for incineration facilities in countries such as Canada. In addition to that, waste incineration was believed to cause significant harm to human health and the environment by local communities [116]. Paper waste incineration produces heat, which can be utilized or further processed to produce electricity. Sometimes waste is incinerated solely for the purpose of disinfection and volume reduction [117]. Waste incineration involves the combustion of solid wastes, releasing gases and energy simultaneously. Incineration of waste can reduce volume by 90% and weight by 70%. While incineration recovers a high amount of energy, it also releases a lot of toxic pollutants such as heavy metals, CO_2, and persistent organic pollutants into the environment [118]. Technologies for waste incineration are developing rapidly. China currently uses three waste incineration technologies, which are stroker grate, fluidized bed, and rotary kiln [114]. The majority of the European incineration plants use moving toward grate technology due to its advantages, such as not requiring shredding or pretreatment, over other incineration technologies, such as the rotary kiln or fluidized beds. However, stroker gate technology has high maintenance costs compared to other technologies. The main challenge in incineration plants is to maintain a constant temperature in the combustion chamber and constant energy output, as increased temperature could corrode or decrease the life of essential plant components [119]. Waste incineration can fulfill the energy demands of smaller states as energy from incineration can be used to generate electricity or power water treatment plants [120].

Among different waste disposal techniques, landfilling is the worst option due to land usage and the release of greenhouse gases. Incineration is considered a better option since much of the energy can be recycled by waste combustion [18]. Among recycling and incineration, the best choice is dependent on factors such as waste handling, loss of quality in paper reprocessing, and energy recovery efficiency. In most cases, incineration coupled with advanced technology is the preferred method of paper waste management [121]. In the past, greenhouse gases and heavy metals released by the incineration process were a major concern. This was solved by the introduction of flue gas treatment systems, which then caused a shift in focus to manage the solid residues of waste incineration [117]. Following the incineration of waste, bottom ash, fly ash, and boiler ash are collected in the form of residues, the majority of which is bottom ash. Bottom ash contains brick, ceramics, and unburned organic matter such as wood, plastic, and fibers [122]. Bottom ash is commonly disposed of in landfills, but it can be recycled to produce construction materials such as concrete and cement. However, due to the leaching of heavy metals and toxic compounds, the recycling or disposal of bottom ash is environmentally concerning [123].

4.4. Nanocellulose

Paper recycling results in the collection of shortened fibers in the form of recycled paper sludge, which is not suitable for paper production. While recycled paper sludge has been used to produce bioethanol, production is limited since bioethanol, in its final form, is a low-cost product [124]. Currently, sludge is disposed of through incineration and landfilling, posing environmental hazards. The conversion of paper sludge into high-value materials, such as cellulose nanoparticles, minimizes the issues related to paper waste disposal [125]. Among these nanoparticles, cellulose nanocrystals (CNCs) are crystalline nano-rods with a thickness of 3–10 nm and a length of a few hundred nanometers. They are usually extracted from pulp fibers using an acid-mediated procedure, which has already been industrialized. They can also be extracted directly from wood and lignocelluloses using a variety of reagents and processes [28,126,127]. Cellulose nanofibrils (CNFs), another form of cellulose nanoparticles, are semi-crystalline spaghetti-like nanoparticles with a thickness of 5–30 nm and a length of a few micrometers. They are usually produced by the mechanical fibrillation of pulp fibers using a wide range of techniques, including microfluidization and homogenization [128,129]. The dimensions and crystallinity of nanocellulose are dependent on its origin and the used method of extraction [130,131]. In addition to their high mechanical properties, biodegradability, and high surface area, CNCs and CNFs are famous for the possibility of modifying their surfaces through abundant hydroxyl groups [131–134]. Due to these interesting properties, CNCs and CNFs have shown great potential in a wide range of applications, including flexible electronics [135–137], energy harvesting materials [138], water treatment membranes [139], drug delivery systems [140–142], tissue engineering [143], biodegradable packaging [144,145], oil clean-up materials [146], and lightweight composites for automotive industries [147]. Nanocellulose is usually extracted from wood pulp and plant fibers. However, several methods have been developed to produce it from paper waste and rejects. The most commonly used acid to obtain CNCs by hydrolysis is sulfuric acid (H_2SO_4) due to its tendency to strongly isolate CNCs and disperse them as a stable colloid system by esterification of their surface hydroxyl groups. Reaction time, temperature, and acid concentration are the main factors that determine the properties of the CNCs [142]. Following acid hydrolysis, CNCs are washed with water to halt the reaction and are then recovered by centrifugation. Due to environmental concerns, acid hydrolysis cannot be implemented in a biorefinery scheme. To overcome this, other routes of obtaining nanocellulose, such as enzymatic hydrolysis, have surfaced. Cellulase is the most commonly used enzyme class for enzymatic hydrolysis due to its ability to attack cellulose fibers with high selectivity [129]. The main drawback of acid hydrolysis is harsh conditions, while enzymatic hydrolysis takes significantly more time. Therefore, subcritical water treatment has been explored because it does not require harsh conditions and an extended period of time [148]. The CNCs and CNFs extracted from paper waste can be used in the papermaking process. CNFs can be utilized as additives to promote adhesion between the fibers and to fill the voids in the paper, which could result in improved strength of the paper in its final form [149,150]. This is because nanocellulose has a large surface area due to its nano-size and many free hydroxyl groups, promoting hydrogen bonding and stronger structure formation [151]. The major issue with the extraction of CNCs and CNFs from paper waste is the calcium carbonate that exists in paper waste. It is expected that it interferes with the extraction process in terms of the formation of by-products and reduces nanocellulose yield and purity [24,152,153]. Still, CNCs and CNFs extracted from paper waste have been explored in a wide range of applications [131,152,154].

4.5. Hydrochar by Carbonization

Paper mill sludge has a high moisture content, which increases drying and transportation costs limiting its utilization. High moisture also attracts microorganisms, which digest the organic fraction of the sludge, resulting in the release of greenhouse gases into the environment [155]. The organic fraction of paper sludge is suitable for hydrothermal carbonization to produce carbon-based materials. Hydrothermal carbonization involves the conversion of wet biomass compounds to high carbon-containing solid products in sub-critical water conditions [156,157]. It usually takes place in a temperature range of 160 °C to 180 °C and self-generated pressure in the range of 1 to 3 MPa and results in a material similar to coal, known as hydrochar. Hydrochar has interesting properties, such as improved hydrophobicity and better dewatering properties compared to paper waste. When dried, its calorific value increases compared to the initial feedstock. Hydrochar can be utilized as solid fuel, low-cost adsorbent, catalyst, and contaminant remover [158,159]. Hydrochar resulting from paper mill sludge carbonization has lower ignition temperature and higher burn temperatures compared to bituminous coal [160]. Hydrothermal carbonization has advantages over other treatments of a similar nature, such as pyrolysis, since it does not require prior drying of feedstock due to water acting as a solvent, reactant, and catalyst [161]. During hydrothermal carbonization, the organic compounds of waste hydrolyze into monomers, which can be turned into hydrochar and organic acids by undergoing several reactions such as dehydration, decarboxylation, condensation, and polymerization [162]. Temperature is the most critical parameter in hydrothermal carbonization, but the type of biomass used also affects the chemical properties of produced hydrochar [163,164]. During hydrothermal carbonization, a fraction of carbon is lost as some of the organic material dissolves in the aqueous phase, and gas is produced in small quantities [165]. The waste undergoing hydrothermal carbonization requires minimal treatments such as dewatering and handling since hydrochar is sterilized during the process [166]. Hydrochar from paper waste can be subjected to a variety of modifications to widen the range of its applications [167,168]. The high ash content in feedstock reduces its suitability for hydrothermal conversion as it contributes to metal corrosion and operating complications in processing plants. In addition, the solid fuel produced by feedstock containing high ash content has low gross calorific value and possesses poor combustion properties. For example, sludge obtained from a primary clarifier at a paper mill is not suitable for hydrothermal carbonization due to the high ash content, whereas Kraft and recycling paper mill sludge has significantly lower ash content, increasing their preference for producing solid fuel [169,170].

4.6. Construction Materials

The increased importance of using environmentally friendly, low-cost, and lightweight materials in the construction industry has resulted in the implementation of waste materials in construction materials since it benefits the environment and maintains construction standards [25]. Therefore, cellulosic fibers obtained from natural sources such as plants are commonly explored for the production of construction materials. However, recycled fibers from paper waste or other sources have not been widely tested, as only a few studies have shown successful implementations of recycled fibers in cement-based composites. Products currently manufactured using recycled cellulosic fibers include plasterboard, insulation materials, and bricks [171]. Paper waste such as newspaper and cardboard can be mixed with cement to produce papercrete, which increases the bonding potential and strength of cement. The production of papercrete involves wastepaper soaked in water to soften the fibers and then added to cement to form blocks or used as mortar. However, there are no standard mixtures of papercrete, which makes it challenging to observe its properties from an engineering perspective. Papercrete production does not require excessive amounts of energy or release by-products that are harmful to the environment [172]. According to Akinwumi et al. (2014), papercrete produced from newspaper demonstrates significantly better structural qualities, such as compressive strength, compared to papercrete produced with office paper. Papercrete has high fire resistance but also absorbs more water. Therefore, it is not recommended to be used for outer walls or walls which are near the ground [173]. A study conducted by Sangrutsamee et al. (2018) investigated the usage of different types of re-pulped paper in cement composite production. Four types of re-pulped fibers obtained from newspaper, office paper, carton paper, and mixed papers were mixed with cement in different ratios, and properties such as compressive strength, bulk density, thermal conductivity, and water absorption were observed. Composites produced from re-pulped carton paper were found to be most effective as they were lightweight and had low thermal conductivity and density. However, they had the drawback of having low compressive strength and high water absorption [174]. A large fraction of inorganic substances in paper mill sludge makes it ideal for the production of several construction materials, such as bricks, cement, and insulation [90]. For example, cellulose fibers mixed with cement and some other additives produce fiber cement products that have high strength, durability, fire resistance, and good appearance. Fiber cement products are used in different construction applications, such as the production of floors, walls, and decorations [175]. The usage of cellulose fibers in construction materials such as cement composites has some drawbacks, such as low durability in alkaline or mineral-rich environments and degradation due to aging mechanisms (mainly alkaline hydrolysis and cell wall mineralization), affecting the reinforcing role of cellulose fibers when used as cement composites [171].

The construction industry consumes clay bricks in large amounts for buildings, and the improvement of these bricks, such as their thermal conductivity, can reduce heat loss through walls. Brickmaking involves the usage of pore-making additives, which help in producing bricks with low thermal conductivity and decreased density while saving clay material. Paper waste can be used as an additive in brickmaking to manufacture bricks, which are lightweight, porous, and have decent strength along with low thermal conductivity [176]. In a study conducted by Goel et al. (2021), paper mill sludge compost was used in brickmaking instead of paper mill sludge due to its low moisture content and ease of shredding. While this concept has not been used until now, this study provides guidelines that can be useful in the future to elevate the concept at a commercial level in brickmaking [177]. Recycled aggregated cement obtained from demolished buildings and paper mill sludge can be used together to produce a control low-strength material, which is not considered concrete or cement but is suitable for backfilling and has certain advantages such as quicker construction time and reduced dependency on equipment due to its self-leveling properties [178].

The incineration of paper waste and paper sludge results in two different types of residues, which are bottom ash and fly ash. Fly ash accounts for less than one-quarter of all residues and contains toxic substances. Bottom ash, however, contains fewer toxic substances in comparison to fly ash and, due to its large solid particles, is suitable for usage in construction. Bottom ash has been previously used as a road filler and for cement production, but it presents a major drawback since it releases heavy metals into the environment [179]. Paper sludge ash can also be used as a clay stabilizer, which was previously done using commercially produced limes or cement. It does not require any energy for production and provides several economic and environmental benefits [180].

4.7. Soil Amendment by Composting

The manufacturing of commercial chemical fertilizers consumes large amounts of resources and degrades the environment due to mineral extraction and calcination [181]. Being an active organic material, paper mill sludge can be potentially beneficial for crops as a source of nutrition [182]. The utilization of paper mill sludge on tropical acidic soils decreases its acidity by neutralizing the soil and increases organic matter and essential nutrients [55]. However, spreading paper mill sludge on agricultural land is challenging, and the impact of physical and chemical properties of organic residues must be tested on soil fertility and site quality. Moreover, paper sludge has high carbon-to-nitrogen ratios and BOD, which causes short-term nitrogen sequestration. In addition, paper sludge is difficult to handle, transport, store, and control the odors released from it. Composting paper sludge minimizes these issues [183]. Composting occurs by mixing the waste with the microorganisms that are naturally found in the soil, such as bacteria, fungi, and protozoa. The microorganisms convert the waste into biologically stable humic substances, which constitute the resultant compost. It is the most advantageous disposal method since it returns the macro and micronutrients back into the ecosystem [184]. Ahmed et al. (2018) studied the biodegradation of different types of papers when composted. Newspaper with ink took the longest (21 days) to degrade when buried in soil, followed by newspaper without ink (18 days). Glossy paper and recycled paper fully degraded the quickest among all four, as they took 14 and 16 days, respectively. The advantages of using paper as compost include the absorption of free water from food materials, reducing odors, and providing a source of carbon, which benefits soils lacking organic matter [185]. Composting paper mill sludge enhances its quality due to the practical physical properties it possesses. Paper sludge compost increases plant growth, and sludge containing calcium carbonate is used to lime soils with lower pH and also provides a source of Zn to marginal soils [186,187]. It is also easily stored without requiring large volumes as compared to raw paper mill sludge. However, the use of paper mill sludge as compost is still not widely welcomed in the agricultural field due to the heavy metals present in paper mill sludge, which can potentially leach into the soil, posing a health hazard [177]. However, a study conducted by Rosazlin et al. (2011) on a compost of recycled paper mill sludge mixed with empty fruit bunches showed that the compost displayed advantageous properties such as no toxicity on plants, 100% seed germination, and high nutrient content. Additionally, the heavy metal concentration in the compost was also within limits [188].

Overall, and as summarized in Figure 3, there are different possible routes for the utilization of paper waste to return its value to the economy and reduce its environmental impact. It is important to mention that some of these routes are also used for the utilization of other kinds of waste, such as electronic waste [189,190]. The greenness and efficiency of the conversion of paper waste into energy are significantly dependent on the form of energy (biofuel, biogas, hydrochar, or heat), the conversion method (SSF, carbonization, anaerobic digestion, or incineration), and the used processing conditions (temperature and others). In terms of the conversion of paper waste into high-value materials such as nanocellulose and soil amendments, the methods vary significantly in terms of efficiency and greenness as they can be chemical, biological, or physical, operating at different severities. In the literature, the comparison in terms of greenhouse gas emissions has mostly focused on the incineration, anaerobic digestion, and composting of different waste streams, including municipal solid waste, food waste, sewage sludge, and others [191–193]. The literature, in general, considers composting to generate less CO_2 emission. Anaerobic digestion comes second, followed by incineration. For example, one study estimated that the greenhouse gas emissions caused by composting municipal solid waste could be around 100 kg CO_{2-eq}/ton waste, compared to around 125 kg CO_{2-eq}/ton waste using anaerobic digestion, and around 1380 kg CO_{2-eq}/ton waste using incineration [194]. These three methods, indeed, are still better options than waste landfilling [195]. In terms of cost and operation, composting is also the cheaper option and the easier to establish and manage [192]. However, several studies have highlighted that the use of an integrated waste management system combining the different methods is the right option to recover the maximum material and energy potential of waste [195].

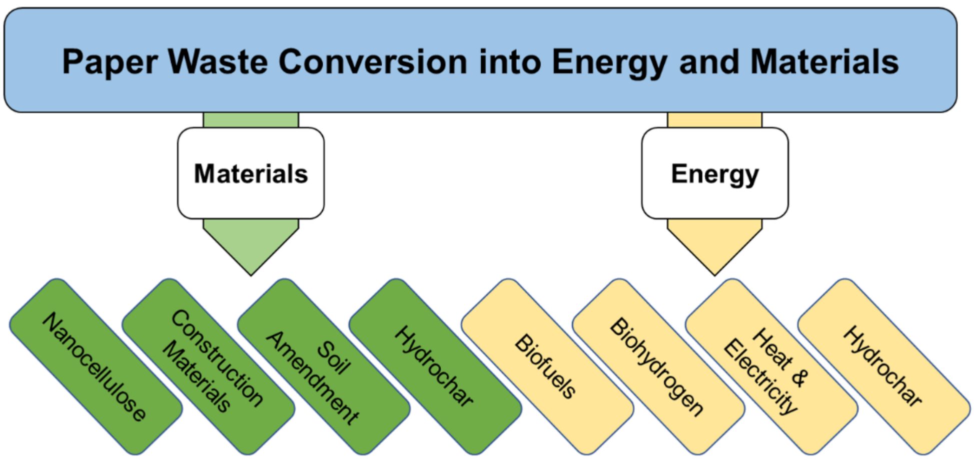

Figure 3. Possible routes for the conversion of paper waste into high-value materials and energy.

5. Conclusions

The pulp and paper industry (PPI) uses massive amounts of natural resources and pollutes the environment, which has raised major concerns, for instance, regarding deforestation. Additionally, the PPI discharges wastewater that can pollute water bodies. Furthermore, the waste produced by this industry includes paper waste and sludge, which require proper management. These wastes are usually burnt or dumped in landfills, which causes a variety of health and environmental issues. Therefore, the recovery aspect of sustainable waste management is of great importance, which is based on a hierarchy of prevention, reuse, recycling, recovery, and disposal. After prevention and reuse, paper recycling is crucial, which significantly lowers greenhouse gas emissions because fewer trees are cut down, and it also lowers manufacturing costs. Paper can be recycled up to seven times, after which the cellulose fibers are considered unsuitable.

The other possible sustainable ways to deal with paper waste emphasized in this review are based on the concept of converting paper waste into energy and high-value materials. Under this concept, biofuel production is possible, as paper is an important source of sugars that can be fermented to bioethanol—an alternate energy source. Other than biofuel production, biohydrogen and biomethane can be produced from paper waste as sources of energy. Heat and electricity can also be generated from the incineration of paper waste. Paper waste and sludge can also be converted into high-value materials minimizing the issues related to paper waste disposal. Paper waste can be used as raw materials for the production of cellulose nanoparticles, which have very advantageous properties and applications. Paper sludge also contains organic fractions that are suitable for hydrothermal carbonization. This process involves the conversion of wet biomass into a high carbon-containing solid product known as hydrochar, which can be used as a solid fuel, low-cost adsorbent, catalyst, and contaminant remover. Another sustainable use of paper waste and sludge is the production of construction materials such as insulation materials, plasterboard, and bricks. The large fraction of inorganic substances in paper mill sludge makes it ideal for the production of construction materials. Finally, paper waste and sludge can be composted for the production of soil amendments, which can decrease soil acidity, increase its organic matter, and provide essential nutrients. The composting of paper sludge or waste that is rich in organic matter can be the most useful disposal method, as this process can return the micro- and macronutrients back to the environment. Overall, the aforementioned processes and possibilities toward the sustainable management of paper waste and sludge have both pros and cons. Therefore, future research should focus on the optimization of the available paper waste utilization processes and on the development of new conversion technologies in order to improve the environmental sustainability of the pulp and paper industry.

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