Available online at http://www.journalijdr.com

IJDR

ISSN: 2230-9926

International Journal of Development Research
Vol. 09, Issue, 06, pp. 28383-28390, June, 2019

RESEARCH ARTICLE OPEN ACCESS

PRODUCTION OF LIGNOCELLULOSIC ETHANOL FROM WASTE PAPER REVIEW ON PRODUCTION TECHNOLOGY

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ARTICLE INFO

Article History:
Received 22nd March, 2019
Received in revised form
03rd April, 2019
Accepted 17th May, 2019
Published online 30th June, 2019

Key Words:

Lignocellulosic biomass, bioethanol, waste Paper, raw material.

ABSTRACT

Waste paper can be found in cities of any country. The possibility of its reuse has moved several researchers to the development of new technologies with the capacity of transforming this waste into economically viable and environmentally sustainable energy that could reduce the use of fossil fuels. The production of ethanol from lignocellulosic biomass from paper waste has been identified as a promising technology with low processing costs and good yield. The objective of this work was to study the ethanol production processes from paper waste, pointing out important characteristics of the biomass and important factors in the technologies available for the production of ethanol from this raw material.

Citation: Ciro Meneses Santos, Philipe Luan. Brito, Andreia Teixeira de Oliveira Santos, Lílian de Araújo Pantoja, Alexandre Sylvio Vieira da Costa and Alexandre Soares dos Santos, 2019. "Production of lignocellulosic ethanol from waste paper review on production technology", *International Journal of Development Research*, 09, (06), 28383-28390.

INTRODUCTION

The great dependence of sources of non-renewable energies and pollutants such as oil and its derivatives point to the need to find alternative sources of energy capable of supplying the demand necessary to support the most varied industrial, agricultural or urban processes that today are powered by fossil fuels. (Zabed *et al.,* 2016, Bensah *et al*., 2015). Current researches signal as a promising technology for the production of fuel ethanol from residual biomasses of lignocellulosic composition (Lin *et al.*, 2012, Aditiya *et al*., 2016). Ethanol is a renewable energy source produced through the fermentation of sugars. Its production is accompanied by great environmental benefits because it's burning results in lower emissions of carbon monoxide (CO), volatile organic compounds, sulfur oxides, etc., when compared to the burning of typical fossil fuels (Jain *et al*., 2014, Soares *et al*., 2002, Crutzen *et al*., 2008). It can be obtained from different types of biomass with significant amounts of carbohydrates, which can be grouped into different categories: sugars, starches or lignocelluloses (Behera *et al*., 2014, Santos *et al*., 2017, Matos *et al*., 2018). The development of technologies for the production of ethanol from lignocellulosic raw materials (agricultural residues, gravels, forest residues, and other low-cost biomasses) has advanced in recent decades. They biomass are abundant sources on the planet and attractive for the production of bioethanol, as well as for the production of several molecules with high added value, such as proteins, microbial, organic acids, enzymes, and biologically active secondary metabolites (Zheng *et al*., 2016, Sun *et al*., 2002, Popa *et al*., 2018). Among the different types of biomass with potential for bioethanol production, stand out short-fiber pulp, such as sludge, paper pulp, and recycled papers. Large amounts of waste paper are produced worldwide. The USA, Japan, China, and the UK discard about 22 million tons annually (Boshoff *et al*., 2016, Nishimura *et al*., 2016). The Brazilian paper and pulp industry have more than 200 industrial units that produce 7.9 million tons of paper (LORA *et al*., Goldemberg *et al*., 2008). These data show us the need for the development of efficient technologies for converting waste paper into economically profitable products, such as bioethanol (Popa *et al*., Nishimura *et al*., 2016, Dubel *et al*.,

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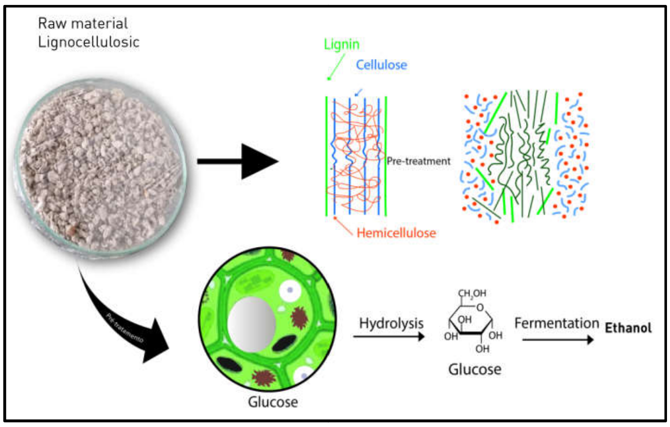
2012, Elliston *et al*., 2013, Sangkharak *et al*, 2011, Wang *et al*., 2011), thereby reducing the large quantities of this industrial waste discarded in landfills, creating environmental problems, such as soil degradation, groundwater contamination, etc. associated with these landfills, could be avoided (Popa *et al*., 2018, Crespo *et al*., 2016). Paper sludge or industrialized vegetable fiber is a potential raw material for the production of second generation bioethanol and is the result of the successive processing of cellulose from the recycling of pulp paper (Byadgy *et al*., 2016, Loelovich *et al*., 2014). The short vegetable fiber is a residue resulting from the reuse of paper chips used in industrial processing for the production of paper towel, toilet paper, napkins, absorbents, among others, with high polysaccharide composition, generated in the recycling units of white paper chips. The characterization of industry residues data, this short fiber has the advantage of having small amounts of lignin and hemicellulose, considerably reducing the cost and time in the pretreatment and hydrolysis stages (Toczylowska-Maminska *et al*., 2017). The bioethanol produced from the use of these short vegetable fibers, besides to minimizing the large quantities generated and discarded in the environment, would still be produced in an economically viable way, due to the reduction of the cost of processing. The result would be the transformation of promising pollutants into clean energy. The production of bioethanol from cellulose requires technologies that overcome technical or economic difficulties (Den et al., 2007, Zheng et al., 2009, Zhao et al., 2008), due to the need to restructure the cell wall, releasing the polysaccharides as a source of fermentable sugars in an efficient and economically way, whose steps basically involve the pretreatment, hydrolysis and fermentation, being a great challenge to find better yields and lower costs (Himmel *et al*., 2009, Mupondwa *et al*., 2017, Canilha *et al*., 2010).

Pretreatment, enzymatic hydrolysis, and alcoholic fermentation

Pretreatment: The available sugars in the lignocellulosic raw material are in the form of polymers (cellulose and hemicellulose), associated with each other and covered by a complex lignin molecule. The disorganization of this complex plant cell facilitates the conversion into fermentable monosaccharides. Pretreatment is the initial stage for the production of lignocellulosic ethanol. aims to disaggregate the recalcitrant material composed of lignin and other polysaccharides available in the plant complex and recover the sugars available in the cellulose and hemicellulose polymers (Trajano *et al*., 2009, Morais *et al*., 2012). These sugars, once released from the cellulose and hemicellulose polymers, undergo yeast action used in alcoholic fermentation. The pretreatment step is fundamental to increase the surface area of the cellulose and reduce its crystallinity and recalcitrance, favoring the process of enzymatic hydrolysis (Mupondwa *et al*., 2017, Guerfali *et al*., 2015, Umagiliyage *et al*., 2015). Figure 1 shows the general scheme of ethanol conversion steps. According to Mupondwa *et al*., (2017) carrying out the hydrolysis of the biomass without the pretreatment step can mean achieving a yield of less than 20% in the process, due to the structural characteristics such as crystallinity, recalcitrance, and porosity of the surface that is covered by lignin and hemicellulose (Zhao *et al*., 2012). Pretreatment can be performed acid pathway and alkaline. In the acid path, the use of an acid catalyst is required. In this case, the conversion is fast, however, through control of the reaction is essential to avoid the formation of undesirable products and inhibitors of fermentative processes, such as phenolic compounds, acetic acid, furfural, and hydroxyl methyl furfural. These compounds may be toxic to yeast. The types of toxic compounds and their concentrations in lignocellulosic hydrolysates depend on the raw material and on the pre-treatment conditions. According to Chandel *et al*., (2007) to analyze the costs of the hydrolysis process, account must be taken of compounds which inhibit the growth of the microorganisms formed by the hydrolysis of the material in question. Several treatments have been used to remove or reduce the concentrations of the toxic compounds present in the fermentation medium, in order to reduce the negative effect of these inhibitors on the hydrolysates. The alkaline pre-treatment can transform the lignin by breaking and destabilizing the connections between hemicellulose and lignin, providing greater porosity to the material (Umagiliyage *et al*., 2015). In this case, chemical pretreatment can be achieved with the use of sodium hydroxide, potassium hydroxide, and calcium hydroxide. These chemical compounds are processed in low temperatures, requiring long times of action, being unnecessary the use of complex reactors (Wan *et al*., 2011). For some types of cellulosic feedstocks, the pretreatment process can be reduced or in some cases even unnecessary due to the low hemicellulose and lignin values, as in the case of paper. In the production of paper by wood pulping the pulping circuit process usually takes place in which digestion and diffusion operations are carried out. The purpose of baking the wood to remove the lignin from the cellulose fibers. The NaOH is one of the most effective alkaline agents used in the lignin removal process in the wood pulping phase, which can promote an increase in the enzymatic digestibility of the material (Chandel *et al*., 2007). The main characteristic of the alkali treatment with NaOH is that it can remove the lignin without affecting much the other components (Himmel *et al*., 2009, Balat e Balat, 2009). In this context, due to the physical-chemical characteristics that paper pulp presents, as a result of its processing, the pretreatment stage can be reduced the costs of bioethanol production (Chen *et al*., Eriksen *et al*., 2008, Tian *et al*., 2014, Araujo *et al*., 2017).



Source: The author (an adaptation of Sousa, 2014)

Figure 1. Schematic representation of the ethanol conversion steps

Enzymatic Hydrolysis: In enzymatic hydrolysis, the catalyst is of biological origin and usually occurs through the use of an enzymatic cocktail composed of cellulases. Cellulases are generally a mixture of various enzymes (Bottino *et al*., 2016) and are classified according to their site of action on the cellulosic substrate, dividing into endoglucanase, which acts in the internal bonds of the cellulosic fiber; exoglucanase, which acts in the external region of the cellulose (Dubey *et al*., 2012, Mupondwa *et al*., 2017). Some factors may affect the enzymatic hydrolysis of cellulose, such as ph and temperature of the hydrolyzed and substrate concentration, the process and reinforce the cellulase activity improves the yield of the hydrolysis (SUN *et al*., 2002, Lemaire *et al*., 2016, Howard *et al*., 2003, Jonsson *et al*., 2016). In the hydrolysis of lignocellulosic biomass, hexoses (D-glucose, D-mannose, D-galactose) and pentoses (D-xylose, L-arabinose) sugars are produced, representing the largest portion of carbohydrates in the lignocellulosic biomass through microorganisms using monosaccharides resulting for the conversion of bioethanol (Rabelo, 2007, Webb *et al*., 1990, Matano *et al*., 2012). The enzymatic hydrolysis process produces a hydrolyzed rich in sugars. However, it is important to observe the costs inherent in the production process so that the final ethanol produced by the lignocellulosic biomass is economically viable. Ligno cellulosic ethanol is mobilizing a growing number of researchers with the objective of increasing the productivity of ethanol lignocellulosic. The aim is to take advantage of cellulose sources as raw material, and the use of low cost and high-efficiency technologies (Den *et al*., 2007, Baek *et al*., 2012, Revin *et al*., 2016). In this context, this work brings together several studies aimed at the production of lignocellulosic ethanol from waste paper, pointing out the production technologies used and the final ethanol yield of each methodology.

Alcoholic fermentation: The alcoholic fermentation takes place after the pretreatment stages with the increase of cellulose digestibility, and later enzymatic hydrolysis for the conversion of cellulose and hemicellulose into fermentable sugars. The next step uses microorganisms to turn the sugars into bioethanol (Sakimoto *et al*., 2017, Bideaux *et al*., 2016). Fermenting microorganisms must have the capacity to generate high rates of fermentation and resist the actions of possible inhibitory products.

The commercial yeast Saccharomyces cerevisiae is used in most of the current fermentation processes for bioethanol production, due to its availability, low cost and capacity to generate high rates of bioethanol fermentation and to have good tolerance to inhibitors (Lemaire *et al*., 2016). However, there is a problem as to the ability of Saccharomyces cerevisiae to naturally metabolize the pentoses, as a result, other yeasts species have been studied, such as Scheffersomyces Pichia Stipitis, Candida Shehatae, and Pachysolen tannophilus, and it demonstrated good results in the conversion of pentoses into sugar (Jain *et al*., 2014, Hahn-hagerdal *et al*., 2007, Kuhad *et al*., 2011, Leolovich, 2014). In short, the conversion of lignocellulosic biomass to ethanol requires microorganisms that efficiently perform the fermentation of the various sugars, since this would allow the optimization of the fermentation process. The summary scheme of ethanol production from lignocellulosic biomass can be seen in Figure 2.

Characteristics features for bioethanol production of lignocellulosic materials and bioethanol from paper: The characteristics of lignocellulosic materials are fundamental for transforming biomass into renewable energy. Data such as high content of cellulose, starch, and hemicellulose are fundamental for this transformation, as well as the low lignin content, which decreases the crystallinity and recalcitrance of the biomasses, favoring the production of these biofuels. The biomass for the conversion process in bioethanol requires an adequate combination of each process step, aiming to achieve a higher yield of bioethanol in an economical and sustainable way. The residue of the paper industry showed excellent results in terms of conversion into ethanol. In addition, the ethanol produced from this raw material contributes to reducing environmental contamination, reducing the amount of waste disposed of, It turns to been a clean and sustainable energy alternative.

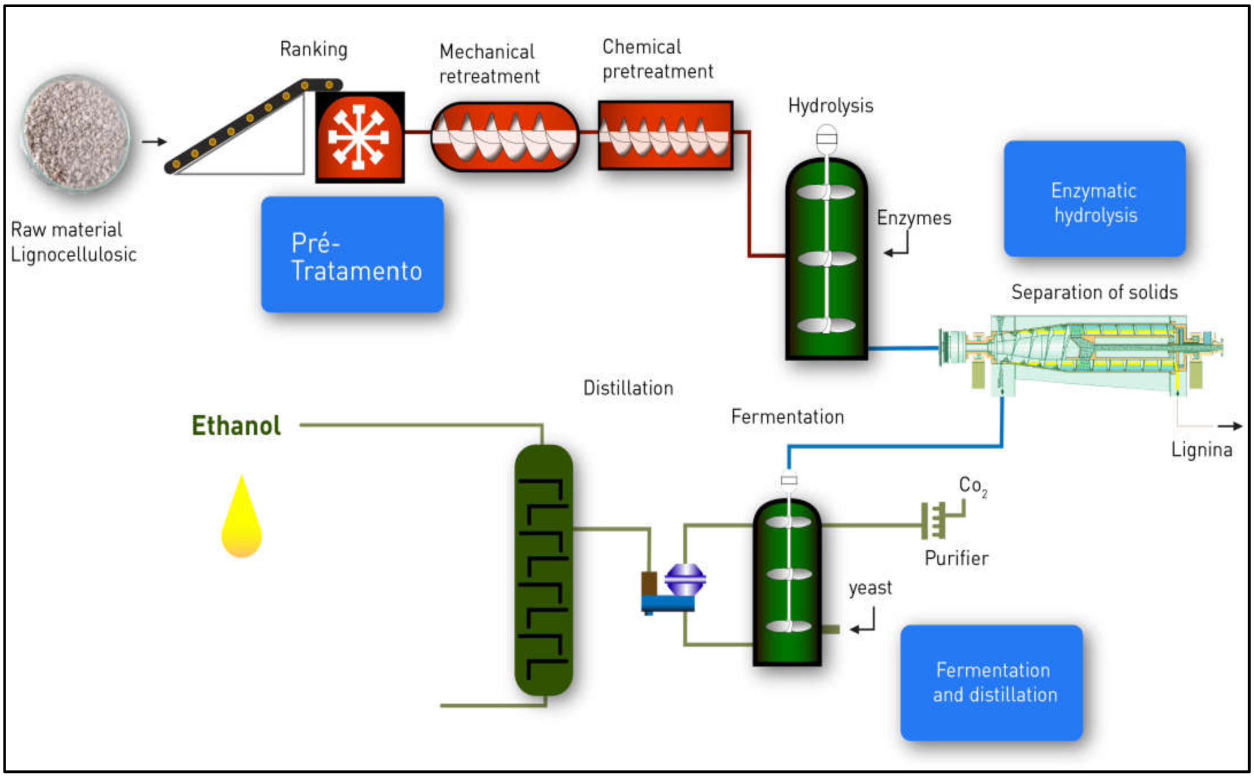


Figure 2. Production of ethanol from lignocellulosic biomass

In this context, in which research and technological developments related to renewable energy sources are emphasized, aiming at solutions that may reduce the impact caused by modern life, and at the same time presented alternatives that can meet the energy needs of the industrial and agricultural. This paper highlights the potential of the cellulosic paper to obtain bioethanol, bringing together the methodologies and results achieved in the production of bioethanol from this raw material. To support this study, a literature review was carried out based on the methodology and technology used for ethanol production, as well as the results obtained, based on articles published in national and international journals, separating those that approached bioethanol production from paper, whereas the most recent, organized into tables for discussions of the results.

Chemical composition of lignocellulosic residues of the articles searched: The characteristics of residues of paper industry presented in the works of Howard et al., Prasad *et al.* (2007), Mosier *et al.*, (2005) Leolovich, (2014), referring the chemical composition of lignocellulosic of different types of wood paper organized in the table 1, with presents the amount of cellulose, hemicellulose and lignin contents, being 38 to 99% of cellulose fraction, 0 to 40% of hemicellulose fraction and 0 to 26% of lignin. These data show us significant amounts of cellulose and low levels of hemicellulose and lignin, indicating the potential of the raw material for the production of bioethanol with reduced processing costs, due to the possibility of reducing or eliminating the pre-treatment step.

Technologies and results found in the literature for the production of ethanol from paper: Several technologies for the production of ethanol from lignocellulosic biomass have been studied in order to reduce processing costs and increase production yield. Table 2 summarizes the work found in the literature for the production of ethanol derived from paper with procedures of pre-treatments, saccharification, and fermentation. Based on Table 2, different types of biomass derived from paper (office paper, recycled paper, pulp, pulp) were used as feedstock for the production of ethanol, using simultaneous and simultaneous hydrolysis and fermentation process saccharification and fermentation. They have shown that they are economically competitive and have good yields in ethanol production (Varotkar *et al*., 2016, Wang *et al*., 2013). Another very important factor observed in paper waste derived from paper mills was the presence of mineral particles, mainly calcium carbonate, which generates inhibitors, preventing the conversion of the cellulose by enzymatic hydrolysis.

Table 1. Chemical composition of Lignocellulosic residue available in the researched literature

Material	Cellulose (%)	Hemicellulose (%)	Lignin (%)	Reference
Wood rods	40-55	24-40	18-25	HOJAMBERDIEV *et al*., 2008
Paper	85-99	0	0-15	Hojamberdiev *et al*., 2008
Newspapers	38-50	15-18	21-26	Hojamberdiev *et al*., 2008
Waste Paper Mill	60-70	10-20	5-10	ROSSI *et al*., 2014
Office Paper	62-87	5-8	1-3	PRASAD *et al*., 2007
Card	61-63	12-13	18-19	MOSISER *et al*., 2005
Packaging Paper	60-73	11-18	7-12	MOSISER *et al*., 2005
Napkin	58-78	6-10	4-5	MOSISER *et al*., 2005
Absorbent Paper	81-84	6-7	3-4	MOSISER *et al*., 2005

Table 2. Technologies and results and results in the production of ethanol from paper

objective	technology	Results	ref.
Evaluates the residue of office paper, ewspapers and paper filter as a microbial substrate for saccharification using wild strains.	Enzymatic saccharification was performed using Aspergillus Sp. Penicillium Sp. Trichoderma Sp. Followed by fermentation (SHF) with yeast Saccharomyces cerevisiae.	The works demonstrate how office paper is susceptibility to hydrolysis by the enzyme Aspergillus sp. With yield (3.76mg ml-1) followed Trichoderma sp. with yield (3.26 mg/ml) and Penicillium sp. with yield (2.6mg/ml).	VAROTKAR*et al*, 2016
Transform short-fiber waste from paper mills into fuel ethanol, attenuate the environmental impact.	Process of enzymatic hydrolysis using commercial cellulases Trichoderme Reesei and SHF fermentation using S. cerevisiae	The study shows the application of a non-ionic surfactant Tween-80 decreased cellulose adsorption and improves the hydrolysis yield.	MIN *et al*., 2015
It evaluates the residue paper hydrolyzed used to ethanolproduction.	Pretreatment acid (H2SO4) 1% v/v, with separate hydrolysis and fermentation (SHF) using P. Stripitis.	The works demonstrate the fermentation of paper hydrolyzate results in the production of ethanol with a yield of 3.73g L-1 with 77% of fermentative efficiency.	DUBEY *et al*., 2012
Evaluate waste paper without pre-treatment used in the production of ethanol.	Enzymatic hydrolysis was performed using Acremonium cellulolyticus followed by (SHF/SSF) using S. cerevisiae.	The study shows the hydrolysis efficiency was 73% and the ethanol yield without pretreatment reached 80% for paper sludge.	GOMES *et al*., 2016
Production of bioethanol as a form of alorization of recycled paper sludge.	Hydrolysis was performed with commercial Celluclast enzymes with enzymatic loading of 20 and 45 FPU g^{-1} and SHF fermentation using S. cerevisiae.	The study demonstrates economic viability of recycled paper slurry, which was easily converted and fermented in 71% and 82% yields and a cost reduction of 40%.	WANG *et al*., 2013
Production of ethanol frompaper sludge using a fermentation substrate capable of fermenting xylose.	Combination of acid pretreatment (H_2SO_4) with enzymatic hydrolysis, with fermentation using yeast capable of fermenting xylose. Using yeasts Candida shehatae, Brettanomyces naardenensis and Pichia stipitis.	The study demonstrates the ability of Brettanomyces yeasts to utilize a wide range of carbon sources and pentoses that grows in limited oxygen and low pH conditions and has yields compatible with Saccharomyces.	JAIN *et al*., 2014
Production of bioethanol using biomass of various recycled waste paper.	Acid pretreatment (H_2SO_4) at 0.5% and enzymatic hydrolyzate with Cellic CTec1 cellulase enzyme using enzyme loading of 7.5 and 16.8 FPU g^{-1}. and separate fermentation using yeast Bacterium Zymomonas.	The analysis shows that bioethanol produced from the waste paper is economically competitive and shows how to increase the recycling of paper and its by-products.	ABDULLAH *et al*., 2015

To reduce cellulose uptake and increase hydrolyzate yield, non-ionic surfactant Tween-80 was used (Min *et al*., 2015, Dwiarti *et al*., 2012).

Papers found in the literature for the production of bioethanol from recycled paper and paper sludge: The recycling and reuse paper sludge or paper fibers is made by complex mixtures composed of organic fibrous materials and chemical additives used in the manufacture of pulp industrial processes (mainly calcium carbonate, kaolinite powder), generated from different process from paper factory, consisting of the steps of sorting, pulping, sieving, cleaning, pickling and bleaching. Its mass consists of 50% of virgin fiber and 50% of recycled fiber, of which 50% is recycled fiber, 25% is composed of fibers from paper chips generated in industries, but not consumed, and 25% from post-consumer shavings [50,66,71,72]. Obtaining ethanol from recycled paper assists in environmental sustainability. Ethanol can be obtained by converting the cellulose present in the recycled paper. In the work carried out by Varotkar *et al*., (2016) were studied the potentialities of recycled paper (office paper, paper filter, and newspaper) to produce ethanol using microbial substrates for saccharification wild strains of Aspergillussp, Penicilliumsp. And Trichodermasp, isolated from different cellulose residues. The cultures were identified according to their characteristics. The temperature and pH parameters were tested and considered suitable for incubation of the substrates used in saccharification. The papers were ground, and a 5g aliquot was withdrawn into the process. This substrate was placed in distilled water and inserted in the autoclaved at 121ºC for 15min. In this study, the office paper showed to be more sensitive to hydrolysis with Aspergillussp, and Tricho dermasp., and demonstrates that it is economically viable to produce ethanol from office paper with an estimated production of 1.3g ml^{-1} to 5gm of office paper. In the study carried out by Guerfali *et al*., (2015) with the objective of exposing alternatives for ethanol production from recycled paper residues, the enzymatic hydrolysis of its cellulose fraction was used, with cellulolytic enzymes produced by Trichoderma Reesei and Aspergillus Niger.

In this work, the researchers use loads of 10 FPU g^{-1} for 48 hours. The production of ethanol from office paper was higher than the tests performed with recycled paper sludge raw material. Looking at the research developed by Dwiarti *et al.* (2012), where Trichoderma Reesei covers are used for cellulose conversion, the results showed a higher yield of ethanol production during the simultaneous SSFsaccharification process. In the work reported by Nishimura *et al.* (2016) for the production of ethanol from recycled paper, the pre-saccharification with Carboxymethyl cellulose enzyme was used, with reducing the simultaneous saccharification time (SSF) and avoiding bacterial contamination of the minimizing the need for high rates of enzymatic loading in the saccharification process with high yield (91.8%) of the ethanol production using recycled paper biomass.In the research developed by Boshoff *et al.* (2016) the objective was to reuse paper sludge as a raw material for the production of bioethanol. In this studypaper pulp samples from several paper mills in South Africa were analyzed. The first result was the differences in chemical composition between pulps from corrugated recycle paper sludge and pulp mills, due to the water retention capacity and viscosity, significantly interfering with the characteristics of the ethanol produced. To determine these characteristics, the researchers classified the samples into categories, such as recycled printed paper, non-recycled printed paper, corrugated recycled paper, and virgin paper, separated according to origin and treatment received during the manufacturing process. The virgin pulp presented the largest fraction of glucose, a lower fraction of ash and consequently better theoretical yield of ethanol, compared to the other categories of raw material researched. In the study by Mendes *et al.* (2016) evaluated the viability of using cellulose sludge and paper from different paper mills as raw material for the production of ethanol. The high polysaccharide content of the sludge and low lignin content indicates excellent biomass for the production of ethanol. The characteristics present in the cellulose sludge significantly reduced the pretreatment process required for raw. In this experiment, the SHF procedure was used for hydrolysis and saccharification.

The work focused on the sterilization and conservation of the batches of raw material for the best yield of the hydrolyzate, and also evaluated the optimal dosage of enzymes to reach the highest yield. No atypical or contaminant or by-product was observed, in the batches of raw material without sterilization, which could reduce time and process costs. However, the lack of sterilization slightly compromised saccharification efficiency. This work also evaluated some scenarios for the optimization of processes with the objective of finding the best yield in bioethanol production. In this case, a 50% reduction in the use of HCl and consequently the need for neutralization with $CaCO_3$ was made. Another hypothesis tested was the efficiency of co-fermentation through the use of yeast (CV-40, Terranel), capable of consuming several types of sugars, such as glucose and xylose. The hydrolysis was performed for 36 hours, with 20 $FPUg^{-1}$ of a commercial enzyme, reaching yield in the ethanol conversion of 49% and yield of $0.78gL^{-1}$.

Conclusion

Second-generation ethanol produced from waste paper presents itself as a good alternative for the production of clean energy. Its physico-chemical characteristics with high carbohydrate and low lignin content favor the production of ethanol. Pretreatment with dilute H_2SO_4 and NaOH showed good results in hemicellulose solubilization. Among the technological processes studied, hydrolysis using the Celluclast® 1.5L and Cellic® CTec2 enzyme cocktail and yeast fermentation Saccharomyces cerevisiae, Trichoderma Reesei and Pichia Stipits indicated the production of ethanol from the paper used with reduced costs, making the process economically and environmentally sustainable. Considering the information generated in the present research, it can be affirmed that the use of waste paper for the generation of ethanol becomes a promising alternative.

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