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# Process development for cigarette butts recycling into cellulose pulp

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## ABSTRACT

Cigarette butts, which are usually thrown on the ground or into ordinary bins, have been recognized as toxic residues since may contain cigarette contaminants and chemicals produced during combustion. Therefore, contaminants in cigarette butts can be leached by rain into surface water and thereby contaminate the environment. In Brazil, according to the National Policy on Solid Waste, all residues must be disposed of in an environmentally friendly manner. Although cigarette butts are not mentioned in the law, due to their characteristics, they may be classified as hazardous waste. At the University of Brasilia, a cellulose pulp production process from cigarette butts has been developed employing alkaline pulping. This process is presented as an alternative to environmentally friendly final disposal of this residue. During the process, a dark liquor is generated, which was found to contain lignin, carbonyls, metals, nicotine and specific tobacco nitrosamines. The dark liquor was treated by acidification to promote lignin precipitation, coagulation with chitosan and  $Al_2(SO_4)_3$  to remove metals and organic compounds and ozonized to oxidize resistant chemicals. The dark liquor presented a high chemical oxygen demand (COD; 29,986 mg/L), which was partially removed by precipitation (20%), chitosan coagulation (66%) and ozonation (45.8%). As the remaining COD was still high, we proposed reusing the clarified effluent in alkaline pulping, which seemed to be the easiest and most efficient procedure with the lowest cost.

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## 1. Introduction

It is known that there are about 5000 constituents in cigarette smoke, which are distilled from tobacco or generated during burning. Among these chemicals, at least 150 of which 44 are present in large quantities in smoke, are recognized as being highly toxic, mainly because of their carcinogenic and mutagenic potential (Hoffmann et al., 1990). To diminish the absorption of these chemicals in the body, as well as their emission into the air, modern cigarettes have cellulose acetate filters, which allow for safer use by reducing or even eliminating those compounds from the smoke. Indeed, these chemicals are partially or fully retained by the cellulose acetate filter, allowing for somewhat safer cigarette consumption.

However, the consumption of cigarettes containing cellulose acetate filters generates tons of toxic cigarette butts annually, which are usually discarded on the streets of cities and are responsible for delivering toxic compounds into soils and limnic water. Indeed, the cellulose acetate filters of cigarette butts are saturated

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http://dx.doi.org/10.1016/j.wasman.2016.10.013 0956-053X/© 2016 Elsevier Ltd. All rights reserved. with toxic compounds, unburned tobacco, ash and paper, and some researchers have highlighted them as environmental pollutants because of inadequate disposal, as butts are typically discarded into ordinary trash and on the ground (Barnes, 2011; Moerman and Potts, 2011; Novotny et al., 2009; Slaughter et al., 2011). Thus, these compounds can contaminate soil and be leached by rain water and brought to surface water and contaminate aquatic environments.

These wastes are small, but are generated in large quantities. For instance, according to the *Anuário Brasileiro do Tabaco*, the consumption of cigarettes in Brazil was about 89.1 billion in 2012 (Poll, 2013). Considering the cigarette butt average weight of 0.4 g, about 35,500 t of this residue is generated annually in Brazil. In the same year, worldwide consumption of cigarettes was about 5.8 trillion, of which about 2.6 trillion were consumed in China, representing about 2,320,000 t of cigarette butts discarded in the environment.

In Brazil, the National Policy on Solid Waste was instituted by Law 12305/2010 (Brasil, 2010), which regulates solid waste disposal and segregation according to the composition of the residue. According to Law 12305/2010, cigarette butts can be classified as municipal and hazardous waste and their segregation and disposal

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must be performed properly in order to prevent soil pollution in garbage dump and consequent pollution of surface and underground water. For this reason, in urban areas, cigarette butts have begun to be collected in special containers. This responsibility may not only be charged upon the consumer or governments. Indeed, some authors underline that "under the environmental principle of Extended Producer Responsibility, tobacco product manufacturers may be held responsible for collection, transport, processing and safe disposal of tobacco product waste (TPW)" (Curtis et al., 2016). However, the final destination of cigarette butts is still a challenge in this field because, until the present work, there has not been a recycling technology available, meaning that cigarette butts are usually incinerated or disposed of in garbage dump.

In order to promote the environmentally friendly disposal of cigarette butts, our research group has developed a technology to recycle them for cellulose pulp production (PI 0305004-1 A) (Costa et al., 2005). The technology consists of separating all the cellulose content in the cigarette butt using four different processes: (i) dissolving the ash and all chemicals adsorbed in the filter; (ii) hydrolyzing cellulose acetate into cellulose; (iii) removing lignin from the remaining tobacco and liberating its cellulose; and (iv) disaggregation of the remaining paper. After filtering the resulting mixture, we obtained a cellulose pulp, which can be used in the paper industry, and a viscous dark liquor, which requires treatment. In the present work, we describe this method of cigarette butt processing, the characterization of the cellulose pulp, the analysis of the dark liquor and its possible treatments.

#### 2. Materials and methods

#### 2.1. Cellulose pulp production

Cigarette butts were collected from cigarette butt collectors located in restaurants, bars and buildings from the Campus of the University of Brasilia and surroundings. The starting mixture contained cigarette butts from different blends and with different amounts of remaining tobacco and paper. Wastes from filter production in the cigarette industry were also used, which contained only cellulose acetate and paper. For the reaction, NaOH was used at a 1% concentration in a water solution to catalyze the reaction. The pulping or cooking process for each fiber (cigarette butts and cellulose acetate) occurred in a period of 3 h, at boiling temperature, in a pilot-scale stainless steel reactor with a 50 L capacity. The reactor was heated with a flame of liquefied petroleum gas (LPG) and the temperature was monitored by a digital thermocouple. A flowchart of the overall process is shown in Fig. 1.

Samples of pulps obtained from cigarette butts and cigarette filter wastes were sent to the Instituto de Pesquisas Tecnológicas (IPT) in São Paulo, Brazil, to evaluate the physical and optical parameters of the cellulose for commercial use as paper. The samples were refined in a Jokro mill for 25 min and the extent of refining was determined by the Schopper-Riegler degree (°SR). In addition, sheets from these samples were processed in TAPPI (Technical Association of Pulp and Paper Industry) forming equipment in order to evaluate the weight, thickness, brightness, tensile strength and elongation. In Table 1 are listed the standard methods used.

#### 2.2. Dark liquor characterization and treatment

Wastewater from cellulose pulp production (dark liquor) using cigarette butts was characterized by measuring the organic matter by determining the chemical oxygen demand (COD), biochemical oxygen demand (BOD) and total organic carbon (TOC). In addition, the settleable solids, the presence of lignin and the content of

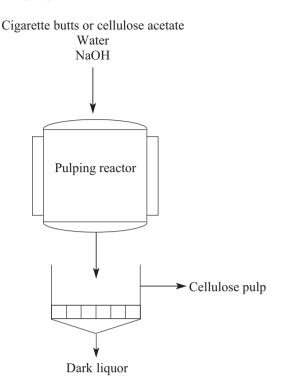


Fig. 1. Pulp production flowchart.

## Table 1

Methods of analysis used on the cellulose pulp.

Analysis	Standard method
Cellulose pulp	
Refining by a Jokro refiner	NBR <sup>a</sup> 14346:1999
Sheet formation	NBR 14479:2000
Paper sheets	
Grammage	NBR 14527:2000
Thickness	NBR 14527:2000
Whiteness	NBR NM ISO 2470:2001
Tensile index	NBR 14527:2000
Stretching	NBR 14527:2000

<sup>a</sup> NBR: standard of the Brazilian Association of Technical Standards.

metal ions and some organic compounds usually found in cigarette smoke and tobacco, such as carbonylated molecules, tobacco specific nitrosamines and nicotine were assessed.

COD determination was carried out by closed reflux colorimetry using a photocolorimeter from Alfakit (model AT10P), in accordance with Standard Methods for the Examination of Water and Wastewater (SMWW), method SMWW 5220D (American Public Health Association, APHA, 1999). BOD determination was carried out by a respirometric method for 5 days at 20 °C (Oxidirect Aqualytic, model Al606), in accordance with SMWW 5210D (APHA, 1999). TOC was determined by catalytic oxidation at 680 °C in a carbon analyzer from Shimadzu (model LCPH/CPN PC), according to SMWW 5310B (APHA, 1999). The samples had been previously filtered through a microfilter Millipore Millex-HV with a porosity of 0.45  $\mu$ m. Settleable solids were determined by the standard method ABNT/NBR 10561/1988 (ABNT, 1988).

The precipitation of lignin from the dark liquor was performed by acidification, as suggested by several studies in the literature by Minu et al. (2012), Mussatto et al. (2007) and García et al. (2009). As the acidifying agents, HCl (6 N), H<sub>3</sub>PO<sub>4</sub> (concentrated and diluted 2% v/v), CH<sub>3</sub>COOH (concentrated) and H<sub>2</sub>SO<sub>4</sub> (diluted 2% v/v) were used. The acidification agent was added to the dark

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