Development of a citrus processing waste-based biorefinery for production of high-added value commodities

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1. Introduction

The global production of citrus fruits is increasing over the years, accounting for 143 million t in 2019 (FAO, 2021). The worldwide industrial processing of citrus fruit is disposing half of its mass as citrus peel waste (CPW) generating 23 x 10^6 t annually, mainly consisting of peels, pulp seeds and segment membranes. Traditional management practices include first generation recycling methods, such as animal feed, composting, disposal in landfills and anaerobic digestion (Tsang *et al* 2019). However, the valuable composition of CPW renders it as a promising raw material for valorization through biorefinery-based treatment. Moreover, significant amounts of wastewater are disposed constituting up to 17 m^3 per t of processed fruit, which burdens citrus processing industries (CPIs). Citrus processing wastewater (CPWW) which includes water for factory cleaning, cooling water, juice concentration and water produced by essential oil extraction, is mainly characterized by large variability of organic loads and other soluble or insoluble compounds, such as sugars, bioactive compounds, essential oils and organic acids (Zema *et al* 2019). Thus, CPWW consists a valuable feedstock, which can be further treated in order to isolate or produce high-added value commodities.

Bacterial Cellulose (BC), which consists a fermentation product, constitutes a biopolymer of significant industrial importance due to numerous unique properties including high crystallinity, high degree of polymerisation, biodegradability, high purity, enhanced mechanical strength and large water holding capacity. These properties increase the industrial interest in various sectors such as food, medical and electronic industries (Hussain *et al* 2019). Previous studies have demonstrated BC manufacture using orange, grapefruit and lemon peels and pulp producing 0.68 g L^{-1}, 6.7 g L^{-1} and 5.2 g L^{-1} respectively (Karanicola *et al* 2021, Cao *et al* 2018, Andritsou *et al* 2018). Furthermore, essential oils as well as bioactive compounds such as polyphenols and carotenoids can be isolated from CPWW as high-added value commodities. Polyphenols consist mainly phenolic acids and flavonoids exhibiting important antioxidant, antiviral, anticarcinogenic, neuroprotective and antimicrobial properties, which could be used in food, pharmaceutical and cosmetic industries (Gomez-Mejia *et al* 2019). Moreover, the carotenoid content could contribute to health as protection from cancer and heart disease providing an important source of vitamin A (Ndayidhimiye and Chun, 2017).

Previous studies have mainly focused on the reduction of the chemical oxygen demand (COD) of CPWW using different treatment methods as anaerobic digestion for methane production (Rosas-Mendoza *et al* 2018) as well as dark fermentation for bio-hydrogen generation (Rosas-Mendoza *et al* 2020). However, given that CPWW constitutes a valuable feedstock for valorization, the present work aims to develop an innovative biorefinery strategy exploiting green technologies for isolation of essential oils as well as bioactive compounds (polyphenols, carotenoids) and production of BC via citrus processing waste valorization.

2. Materials and methods

CPWW was initially characterized through standard analytical methods. The physicochemical characteristics assessed included total sugars (TS), total phenolic content (TPC), COD, total solids and free amino nitrogen (FAN). Essential oils quantification was performed using GC-FID following extraction using hexane.

The phenolic content entailed in the liquid fraction of CPW has recovered using nonionic resins and biochar as adsorption materials, which are capable of adsorbing polar and non-polar polyphenolic compounds, in batch experiments. The desorption capacity of each material was assessed using packed columns at different flowrates of ethanol.

The liquid fraction remaining following essential oils and bioactive compounds isolation was employed in BC fermentations using *Komagataeibacter sucrofermentans* DSM 15973.

3. Results and Discussion

A biorefinery approach was used for isolation of high-added value commodities (bioactive compounds and essential oils) and production of bacterial cellulose using CPWW emitted from the industrial process. CPWW constituted 104 g L^{-1} COD, 57.3 g L^{-1} TS, 1.3 g L^{-1} TPC (gallic acid equivalents), 98.3 mg L^{-1} FAN, 357 mg L^{-1} d-limonene and 5.5% of total solids which are rich in bioactive compounds and fibre content.

The present work mainly focused on isolation of polyphenols from the aqueous stream through adsorption evaluating different types of resins, effluent volumes and polyphenols desorption. Specifically, polyphenols adsorption reached 75% using a non-polar resin, while 94% of polyphenols desorption was achieved at an ethanol flowrate of 0.1 L h^{-1}. The remaining sugar-rich liquid was employed in *Komagataeibacter sucrofermentans* fermentations yielding up to 0.26 g_{BC} g_{TS}^{-1}.

4. Conclusions

An innovative biorefinery strategy was developed for bioactive compounds recovery and BC production exploiting the waste streams emitted from CPIs.

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