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Review

Citrus waste as feedstock for bio-based products recovery: Review on limonene case study and energy valorization

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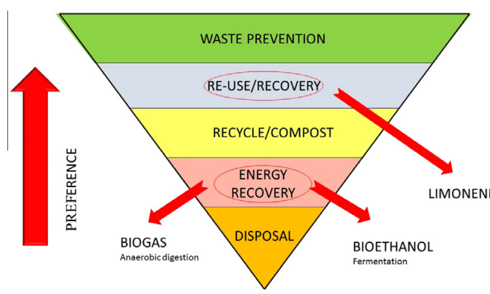
HIGHLIGHTS

- Citrus peel and fruit juices waste streams are rich in limonene molecule.
- Limonene can be used in the nutritional, pharmaceutical and cosmetic fields.
- Traditional techniques to recover limonene involve the use of polluting solvents.
- Cost-benefit analysis is required to estimate the most viable extracting technique.
- Anaerobic digestion and fermentation are suitable post-limonene extraction processes.

GRAPHICAL ABSTRACT



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ABSTRACT

The citrus peels and residue of fruit juices production are rich in D-limonene, a cyclic terpene characterized by antimicrobial activity, which could hamper energy valorization bioprocess. Considering that limonene is used in nutritional, pharmaceutical and cosmetic fields, citrus by-products processing appear to be a suitable feedstock either for high value product recovery or energy bio-processes. This waste stream, more than 10 MTon at 2013 in European Union (AIJN, 2014), can be considered appealing, from the view point of conducting a key study on limonene recovery, as its content of about 1% w/w of high value-added molecule. Different processes are currently being studied to recover or remove limonene from citrus peel to both prevent pollution and energy resources recovery. The present review is aimed to highlight pros and contras of different approaches suggesting an energy sustainability criterion to select the most effective one for materials and energy valorization.

Contents

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1. Introduction

Landfill disposal of food waste is currently only allowed if a waste has previously been subjected to a resource and energy recovery process. EU directive 2008/98/EC establishes that citrus waste, like any other kind of food waste, cannot be disposed of in a landfill without a previous valorization, because of environmental and economic concerns of landfill technology. The disposal of food wastes leads to different problems of both economic and environmental nature, due to its high fermentability, high transportation costs, lack of disposal sites and the difficulties to store for long time organic wastes. The goals fixed by the EU are very challenging and encourage to move toward high recycling targets, paving the road from a linear economy to a circular economy as a real answer for the challenge of globalization (EU, 2014). According to the food waste hierarchy (Papargyropoulou et al., 2014), the first level of attention is directed toward the need to prevent the formation of waste; the following next steps concern the reuse or recovery and recycling of suitable materials and afterwards the energy recovery through a thermochemical or biological process; only at the end, when there are no more alternatives, it is allowed the disposal of residuals into a landfill.

The goal of a correct waste management is not only to reduce the disposed waste volumes, but also to make use of it in various ways. Citrus waste can in fact be considered as a very important renewable resource for biofuel production, and the research in this field has been driven by both environmental and economic aspects (Mamma and Christakopoulos, 2014). Eighty-nine million barrels of crude oil were consumed per day throughout the world in 2013, and the consumption of liquid fuels for transportation, including food transportation, has been forecast to increase by 57% by 2040 (IEA, 2015). The dependence of fuel on fossil fuel is a global issue, since the resources on the Earth are limited. In fact, British Petroleum, in its latest "BP Statistical Review of World Energy June 2015" (BP, 2015) forecasts that total world proved oil reserves of 1700.1 billion barrels at the end of 2014 is sufficient to meet 52.5 years of global oil production of the same year; this prediction, joint to the Global Warming effect due to the use of fossil resources, gives the idea of the urgency to find different sources to fueling the transportation of goods and peoples. In this panorama, obtaining biofuels from citrus waste would allow some beneficial effects because of the use of renewable sources with low contribution to the production of greenhouse gas. Moreover, this would prevent acid rain, due to the combination with sulfured compounds from fossil fuels. Instead, biofuels, such as bio-ethanol or biogas, can be considered an innovative alternative to traditional fuels and they could help to prevent all the negative effects of petroleum transportation fuels. Citrus waste could be used as a suitable substrate for fermentative processes, thus complying with the waste-to-energy principles.

Citrus waste is usually dried in order to use it as animal feed or for pectin extraction (Mamma et al., 2008). However, a high energy cost dehydration process is necessary to dry and pelletize the citrus waste for animal feed; this disadvantage, together with the low nutritional properties and the bitter flavor, implies that large quantity of juices production waste is dumped into landfills, thus resulting in the loss of a great potential resource. In order to maximize the exploitation of this available resources, citrus peels should be subjected to a two-steps recovery of matter and energy. In this way, the recovery of matter offers a two-fold advantage: limonene, a high-added value molecule, is obtained and a metabolite that is dangerous for the fermentative process is removed. Anaerobic digestion (AD) or ethanol fermentation can be optimal choices for organic waste valorization and renewable energy production.

The aim of this work is to describe the possible achievable techniques for the recovery of materials and energy, analyzing for each of them advantages and disadvantages, in order to up-to-date the available information to suggest the way toward the most appropriate treatment to manage the citrus waste in the perspective of global sustainability of the use of resources.

2. Citrus waste characterization

The most abundant tree crop is citrus fruit, with a world-wide production of more than 88 million tons per year; orange is the most abundant (about 80% of the total) in this large category, which also includes grapefruit, lemons, limes and mandarins (Marín et al., 2007). According to FAO (2014), the most important orange producing country is Brazil, followed by USA, with 7.5 million tons of oranges (of which 70% are farmed in Florida) and by China, with a rising trend of yearly 7 million tons of oranges; oppositely, EU shows a downward trend, with less than 6 million tons of oranges (AIJN, 2014). The third most abundant citrus fruit, after orange and mandarin, is lemon, whose word production is about 4.2 million tons, and is concentrated in Argentina, Italy, Spain, USA and Mexico (FAO, 2014). About 50% of the quantity of this fruit is processed for fruit juice and marmalade production and approximately 50–60% w/w of the processed fruit becomes waste (Wilkins et al., 2007a). In US alone, over the 2008–2011 period, 21.16 million tons of citrus fruit (especially oranges and grapefruit) were processed, which resulted in a solid waste generation of about 10 million tons (NASS, 2011). This waste consists of: (1) peel and pulp, (2) fruit that has not been processed because it was damaged and/or did not conform to quality standards and (3) returned surplus goods. After the production of orange juice, the waste is composed of 60–65% w/w of peels, 30–35% w/w of internal tissue and the remaining of seeds (Crawshaw, 2003).

Citrus waste is voluminous, heterogeneous, chemically complex and highly biodegradable, with a COD of 1085 mg O_2/g in case of orange peels (OP) (Siles et al., 2016). Moreover, it is characterized by a low pH (3–4) and a high organic content, more than 95% w/w of the Total Solids (TS) (Ruiz and Flotats, 2014). For these reasons,