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Review

Recent advances in the processing of green tea biomolecules using ethyl lactate. A review

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A B S T R A C T

Background: Green tea and its decaffeinated products are foodstuffs which involve a great commercial market due to their interesting healthy properties. Catechins are the main phenolic compounds in tea leaves and these are receiving considerable interest due to their potential benefits on human health. In this sense, many studies have been performed with the aim of removing the caffeine and obtaining either decaffeinated green tea leaves preserving the catechins or decaffeinated catechins-rich extracts.

Scope and approach: In this review, different methods and solvents described in the literature for green tea decaffeination and recovery of high valued catechins are revised. Particular attention is given to ethyl lactate, an agrochemical green solvent studied by the authors for the extraction of caffeine.

Key findings and conclusions: A diversity of results has been reported for the different green solvents and extraction techniques studied to remove caffeine from green tea leaves and extracts in the last years. Nevertheless, despite the solvent used, the loss of catechins is unavoidable to some extent. In this sense, ethyl lactate has demonstrated higher selectivity and efficiency with respect to commercial and other agrochemical solvents currently used to that end. Additionally, combining ethyl lactate with Supercritical CO_2 Anti-Solvent technique, a decaffeinated green tea precipitate could be obtained, overcoming limitations of presently studied procedures to obtain decaffeinated extracts and demonstrating to be a selective and effective alternative for catechins purification.

1. Introduction

Tea is a popular beverage traditionally used as a drink and medicine in the southern regions of Asia. On account of its healthy properties, the consumption of tea has become widespread in the western countries. Additionally, the stimulant effect of caffeine and the prevalence in these countries of chronic diseases, such as hypertension, has promoted the increment of decaffeinated tea market. Based on this, many studies have been carried out with the aim of finding new solvents and/or extraction methodologies that allow obtaining decaffeinated tea with an improved quality in an efficient manner, as well as decaffeinated tea extracts, enriched in phenolic compounds, which can be used as a nutraceuticals and food ingredients.

Chlorinated solvents have demonstrated to be the most selective solvents for extracting and fractionating caffeine. Nevertheless, the growing environmental problems and the adverse health effects of these solvents have influenced in the demand of efficient solvents, more environmentally sustainable and safe, according to the Green Chemistry concepts (Anastas & Kirchhoff, 2002). Regarding green solvents, it is important not only to use non-toxic, safe and environmental friendly solvents, but also to use solvents which production follows the principles of Green Chemistry (Anastas & Warner, 1998).

Ethyl lactate is an agrochemical solvent defined as GRAS (Generally Recognized as Safe) and due to its low toxicity it has been approved by FDA (Food and Drug Administration) and EFSA (European Food Safety Authority) as a pharmaceutical ingredient and food additive. Ethyl lactate has interesting properties, such as being a non-corrosive, non-carcinogenic, non-teratogenic, biodegradable and non-ozone depleting solvent.

Ethyl lactate has demonstrated to be a more selective solvent for the extraction of caffeine from green tea leaves than water and organic solvents studied and used at industrial scale until now (Villanueva Bermejo, Mendiola, Ibáñez, Reglero, & Fornari, 2015). Added to CO_2 as a cosolvent and using the same conditions as

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commercially, a higher caffeine extraction rate in a shorter extraction time period was also achieved (Villanueva Bermejo, Ibáñez, Reglero, & Fornari, 2016). Additionally, using this green solvent, a dry decaffeinated extract with high concentration of catechins and phenolic compounds that can be used as food ingredient or nutraceutical, can efficiently be obtained (Villanueva Bermejo, Ibáñez et al., 2015). Therefore, the drawbacks of the other methods recently or traditionally studied for this purpose, such as liquid-liquid partitioning, membrane technology or the use of adsorbents, can be solved.

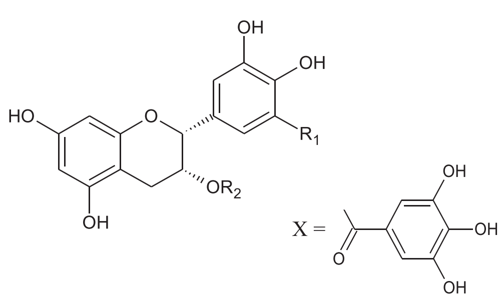
In this review, the different methods and solvents reported in the literature for the decaffeination and recovery of high valued catechins are revised. In this regard, Section 2 describes in detail the bioactive compounds present in green tea leaves, emphasizing the relevance of caffeine and catechins due to their bioactive properties and high content in this matrix. Section 3 describes the main characteristics of the green solvent ethyl lactate and delves into the use of this solvent that has been reported for food applications until now. The following two sections are focused on the extraction methods and solvents currently performed for obtaining caffeine and catechins from green tea, contrasting the results with that reported for ethyl lactate. On the one hand, Section 4 inspects the most recent works related to decaffeination of tea leaves. On the other hand, Section 5 shows the latest studies performed to improve the extraction of caffeine and catechins from green tea leaves and to fractionate and concentrate these compounds from the obtained green tea extracts, applying new and traditional extraction and purification technologies. Finally, general remarks regarding the use of ethyl lactate as a new green solvent for the decaffeination of green tea leaves and green tea extracts are pointed out.

2. Green tea composition and bioactive compounds

Tea is the beverage prepared from the leaves and shoots of *Camellia sinensis* plants which belong to *Theaceae* family, the major varieties being the sinensis (*C. sinensis* var. *sinensis*) and the assamica (C. *sinensis* var. *assamica*). The brew prepared from the plant is one of the most consumed beverages in the world, mainly owing to its organoleptic properties and stimulant effects and in recent years has been intensely studied due to its potential health benefits. It has resulted in an increase in tea consumption in Europe and United States over the last 20 years (Engelhardt, 2010). Today tea is consumed, not only as beverage, but also as an ingredient to elaborate other food products, such as canned drinks, confectionery products, ice creams and cosmetic and phytotheraphy products (Kawakatsu, Kobayashi, Sano, & Nakajima, 1995; Shi & Schlegel, 2012). Variations of the processing result in the different types of teas (green, black, white, oolong and pu-erh) specially the so-called fermentation that is in fact an enzyme conversion of some constituents (mainly flavonols) by leaf enzymes. In the case of green tea, it has undergone minimal oxidation during processing. For this reason, its composition and organoleptic properties differ from the other types of tea (Wan, Li, & Zhang, 2009). Unlike fermented teas, green tea is a plentiful source of phenolic compounds which can reach up to 30% (dry weight) of the leaves (Shahidi & Naczk, 2004; Shi & Schlegel, 2012). Regarding phenolic compounds, flavonols and flavones (mainly in the form of flavonol and flavone glycosides, such as kaempferol, myricetin, quercetin, apigenin and luteolin glycosides), are present in the tea leaves, as well as phenolic acids such as 5-O-galloylquinic. These compounds are preserved during oxidation so that its concentration can be similar to that in other types of tea, whereas the concentration of other phenolic compounds considerably changes. This is the case of catechins monomers, the major phenolic compounds contained in green tea leaves (Engelhardt, 2010).

Catechins (flavan-3-ols) are the most abundant phenolic compounds in tea leaves (7–25% dry weight) and in some cases, they could represent 90% of the total mass of phenolic compounds (Shahidi & Naczk, 2004; Wei et al., 2011). Over the oxidation step, catechins polymerization and oxidation is produced, therefore green tea contains greater amounts of these compounds than teas subjected to oxidation. Catechins contribute to astringency and bitterness and they are one of the most bioactive compounds of the tea leaves (Chaturvedula & Prakash, 2011). In terms of stereochemistry of the 3′,4′-dihydroxyphenyl group and hydroxyl at position 3 of the C-ring, catechins are divided in trans-catechins and cis-epicatechins. At the same time each of them can be present as two optical isomers, (+) and (-)-catechins/(+) and (-)-epicatechins, respectively. Catechins can be esterified with a gallic acid molecule at position 3 of the C-ring giving rise to catechin gallates. The main catechins forms (see Fig. 1a) in green tea are (-)-epicatechin (EC), (-)-epigallocatechin (EGC), (-)-epicatechin gallate (ECG), and (-)-epigallocatechin gallate (EGCG). Of them, EGCG is usually the

(a)



R^1	R^2	Compound
H	H	Epicatechin
OH	H	Epigallocatechin
H	X	Epicatechin gallate
OH	X	Epigallocatechin gallate

(b)

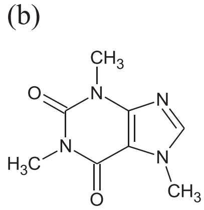


Fig. 1. Chemical structure for the main catechin monomers (a) and caffeine (b) present in green tea.