Valorization of Ellagic Acid from Chestnut Shell as a Natural Anti-anxiety Food Supplement

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Abstract
Mental health is on the agenda, especially in pandemic times due to the COVID-19 virus. Nowadays, psychological problems, such as anxiety and depression, have been more discussed and have become even more critical during lockdown. This paper proposes the development process of an innovative product whose active ingredient - ellagic acid, extracted from the chestnut shell - has anxiolytic properties and can be potentially used in consumers’ daily lives, preventing this type of illness. The product presented, *Do Nut Stress – Acid ellagic enriched pancakes for anxiety control*, consists of a pancake powder, considered a healthy and nutritious meal.

Author Keywords. Anxiety. Depression. Ellagic Acid, Pancake. Chestnut.

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1. Introduction
The chestnut tree is one of the most abundant fruit trees in northern Portugal. Its fruit, the chestnut, is highly sought after worldwide and extremely rich in nutrients. This work explores an innovative strategy to take advantage of the chestnut production residues as raw materials to foster industrial activities.

In-depth research of the chemical potentialities of the several parts of this tree was conducted, from the leaves to the fruit; it was found that chestnut extracts contain a high number of phenolic compounds, either from the chestnut shell, leaves or flower (*Braga 2014*). Within these phenolic compounds, the ones that are highlighted are tannins, ellagic acid, chlorogenic acid and rutin. Those compounds have a high antioxidant function (*Evtyugin, Magina, and Evtuguin 2020*).

According to the properties of the main components of chestnut extracts, several ideas were put on the table to meet current market needs. This process resulted in a limited selection of ideas choosing the one that better fulfilled that needs.
Ellagic acid is a natural polyphenol; it is present in a variety of fruits and vegetables (Sepúlveda et al. 2011), with large content in the chestnut shell. This compound has been proven to have anti-anxiety and antidepressant-like activity in mice, likely because of its interaction with adrenergic and serotonergic systems. These characteristics can be explored and possibly used to prevent psychological illnesses (Dhingra and Chhillar 2012). Ellagic acid shows anti-mutagenic, antioxidant, anti-tumor, antimicrobial, antibacterial, and anti-anxiety properties (Sepúlveda et al. 2011; Evtyugin, Magina, and Evtuguin 2020). Also, this compound is believed to have anti-cancer properties due to its ability to inhibit the activities of DNA topoisomerases that are involved in carcinogenesis. Ellagic acid has been studied for its beneficial health effects.

Over the last few years, there has been a growing concern about increasing diseases associated with mental health. One of the biggest concerns is stress-related disorders since stress can increase the risk of mental diseases, such as anxiety and depression, and suppress the immune system. In Portugal, anxiety caused by stress is present in the daily life of thousands of Portuguesees, being the main cause of work-related stress. In this work, after selecting the idea, more narrow research could begin to reach the final objective: to design a new and up-to-date product with the potential to be launched on the market.

The research on extraction methods focused on the extraction of ellagic acid from the chestnut shells; the product manufacturing process and a brief economic analysis were mandatory steps and are described in detail throughout this work.

A final product culminated: Do Nut Stress, a pancake powder enriched with ellagic acid to help control anxiety levels, targeting the youngers as the main potential buyers.

2. Raw Material

The European chestnut (Castanea sativa Mill.) belongs to the Fagaceae family and the genus Castanea (Costa 2018b). This tree can be explored for various purposes; however, the most common are wood and fruit production. According to information dating from 2010, Portugal has the 5th largest harvest area and it is the 7th largest chestnut producer (Costa 2018a).

Based on the available information and productivity data, the production of chestnuts in Portugal was around 34 thousand tons per year, with a cost of 2.8 thousand euros per ton from local producers. Most of the chestnut trees area distribution is located around Bragança, Vila Real and Viseu (INE 2019).

According to the latest figures of the Corporate Statistics Database of the Food and Agriculture Organization, in 2010, the registered area of chestnuts was 34 thousand hectares, with a target of 45 thousand hectares planted by 2022, assuming densities between 70 to 100 trees per hectare (Fórum Florestal 2020).

In national and international markets, European chestnuts are perceived as expensive. This high value is easily explained, considering that chestnuts are gluten-free and have super organoleptic properties, essential amino acids, quality lipids, and considerable amounts of minerals and fibers. European chestnut has a promising future lying ahead, as this resource’s growth has been verified throughout the last decades for the production of traditional products and new products associated with natural and organic foods (Costa 2018a).

Chestnut shell is a by-product that contains several bioactive ingredients, such as flavonoids and phenols. An et al. (2021) propose a process of extraction and recovery of ellagic acid from waste chestnut shells, reaching a purity of ellagic acid in the extracts of 85.6%.
3. Chemical Product Design (CPD)

Regarding innovative products, controversy has raged among economists and scientists about the relative significance of “technology push” versus “market pull” theories. While technology push is related to the desire to valorize disruptive technologies, the market pull is guided by market needs. In the latter, research staff develops new products in an effort to respond to customer problems or suggestions. Therefore, the development of a new product requires extensive research on how, in this situation, the active principle can enhance a product, always aiming for the market needs.

3.1. Needs & Ideas

After the study of the chestnut tree’s active principles and their corresponding properties, it was possible to distinguish two different application areas, namely Health and Cosmetics.

For the rutin active principle, it was found that it can improve Vitamin C absorption, potentiating healing properties (Bittar and Leite 2018), enhancing sun protection factor (SPF) in sunscreens when associated with physical filters like titanium dioxide (TiO₂) and zinc oxide (ZnO) (Alencar Filho et al. 2016), and function as an antioxidant in cosmetics because of its biocompatibility with human skin and maintenance of collagen in healthy conditions (Brand-Garnys et al. 2007). Chlorogenic acid is proved to be an antibacterial principle against Gram-positive and Gram-negative bacteria (Lou et al. 2011). On the other hand, ellagic acid has antidepressant and anti-anxiety properties (Dhingra and Chhillar 2012).

After identifying the properties of the active compounds, possible products to put on the market were brainstormed. Rutin could be used as a component of a mouthwash formulation or in a sunscreen. On the other hand, chlorogenic acid may be incorporated into disinfectant wipes and anti-ageing creams to clean surfaces and also enhance skin rejuvenation, respectively. An innovative pancake powder mix may be formulated with ellagic acid, mainly targeting young people struggling with anxiety. At last, quercetin may be incorporated into a shampoo for fragile hair. Table 1 lists the main ideas.

| Product          | Scientific Maturity (35%) | Innovation (25%) | Ease of Engineering (20%) | Cost (20%) | Total  |
|------------------|---------------------------|------------------|---------------------------|------------|--------|
| Mouthwash        | 2                         | 4                | 4                         | 3.5        | 3.2    |
| Disinfectant wipes | 4                         | 4                | 4                         | 3.5        | 3.9    |
| Pancake powder   | 4                         | 5                | 5                         | 4          | 4.5    |
| Sunscreen        | 4                         | 2                | 4                         | 3          | 3.3    |
| Anti-ageing cream| 2                         | 4                | 4                         | 3          | 3.1    |
| Shampoo          | 2                         | 4                | 4                         | 3          | 3.3    |

Table 1: Selection matrix for new applications of the active principles extracted from chestnuts shell

3.2. Selection

The previous ideas were evaluated through a selection matrix (Table 1), where they were classified according to 4 criteria, with the following weighting: scientific maturity (35%), innovation (25%), ease of engineering (20%) and cost (20%). The criteria have been ranked from 1 to 5, where the value of 1 is attributed to the worst rating and the value of 5 to the most favorable.

According to the authors’ know-how, it is considered the highest weighting to scientific maturity; this parameter considers the published articles that refer to the effect of the active ingredient in the corresponding product. To follow, it considered the originality and innovation compared to the ones already existent in the market. The ease of engineering, i.e.,
the complexity of the manufacturing processes and the estimated cost of production, was the same weight select the idea.

The product that obtained the highest score was the pancake powder, which stood out mainly for its innovation and ease of production.

Anxiety is related to physical changes like increased heart rate, and it is associated with worrying thoughts. The feeling of anxiety is natural, but it becomes a disorder when it occurs regularly and disproportionately. This is a very current issue, but some researchers suggest that anxiety is not increasing, but rather people are more open to accepting this disorder and treating it (Shapper 2020). In any case, this problem can really interfere with a person's daily life, especially in the lives of students and professionals with great responsibility.

So, the selected product was the pancake powder containing ellagic acid, which has an anxiolytic function, and it was mainly designed for students, although anyone can consume it. Indeed, pancakes are a very attractive food product for the consumer. They have appeared from the most original to more healthy recipes.

Thus, a composition of pancake powder with ellagic acid has been developed, which can bring some benefits to consumers' mental health.

In addition, with the growing dietary restrictions, the presented product still has two special and differentiating characteristics: gluten and lactose-free, making it more inclusive.

This product consists of a powder based on flour, milk powder and sugar, together with ellagic acid, prepared to mix with 50 mL of water or milk and two eggs (optional) and to cook in a frying pan. All the ingredients and their quantity are presented in Table 2. The amount of ellagic acid required takes into account the minimum required determined by Girish et al. (2013) to produce substantial effects on anxiety (in mice). It is noteworthy that in this work, there are no experimental tests carried out.

| Ingredient         | Quantity (% wt.) |
|--------------------|------------------|
| Wheat flour        | 79.01            |
| Sugar              | 6.93             |
| Milk powder        | 6.58             |
| Baking powder      | 4.79             |
| Vanilla extract    | 1.61             |
| Salt               | 1.05             |
| Ellagic acid       | 0.03             |

Table 2: Composition of pancake powder

4. Manufacture

4.1. Extraction process

A method studied by An et al. (2021) is highlighted to understand the ellagic acid extraction process from the chestnut shell. The current process uses ultrasound-assisted extraction (UAE) technology with choline chloride/n-propanol deep eutectic solvent (DES). It is possible to obtain an extraction value for ellagic acid of 4.64 mg/g of chestnut shell with a high recovery percentage of 94.9 %.

Chestnut shells were dried, pulverized, and sieved. Meanwhile, the DES was synthesized using choline chloride and n-propanol with a molar ratio of 1:1 in an oil bath at 80 °C. Afterward, the extraction solvent was added to chestnut shell powder and extracted by an ultrasonic cleaner with an extraction power of 200 W for 70 min. The extracted solution was centrifuged, and water was added to the supernatant to precipitate the ellagic acid. The mixture was left in an ice bath at 0 °C for 6 h, centrifuged and then dried to obtain high purity ellagic acid.
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(85.6%) (An et al. 2021). Figure 1 shows the process flow diagram to produce Ellagic acid from chestnut shell.

![Process flow diagram to produce Ellagic acid from chestnut shell](image)

Figure 1: Process flow diagram to produce Ellagic acid from chestnut shell

4.2. Manufacturing process
After ellagic acid extraction from chestnut shell, it is necessary to add all the ingredients, wheat flour, sugar, milk powder, baking powder, vanilla extract, salt and ellagic acid in a blender (Pedrick and Wallis 2004), which is the first step of the manufacturing process. Afterward, the product is sterilized and packaged. Finally, the packages are labeled, and the product is ready to be distributed.

5. Economic Analyses
The economic analysis was made for a period of 5 years, from 2021 to 2025. The costs associated with the enterprise were estimated, such as raw materials, supplies, staff costs, investment costs, and financing. The total initial investment estimated was 810 thousand €. It is to highlight that this analysis is a rough estimate.

It was considered production of 1.31 million packed for the 1st year, followed by an increase of 10, 12.5, 15 and 17.5%. In order to determine the price of the product, it is necessary to assess the ingredients’ costs. Table 2 shows the prices of the ingredients.

| Ingredient         | Price (€/kg) | Price per package (€/package) |
|--------------------|--------------|------------------------------|
| Wheat flour        | 2.23         | 0.880                        |
| Sugar              | 0.33         | 0.011                        |
| Milk powder        | 1.65         | 0.054                        |
| Baking powder      | 1.57         | 0.038                        |
| Vanilla extract    | 1.44         | 0.012                        |
| Salt               | 0.07         | 0.002                        |

Table 3: Ingredients prices

Moreover, for the extraction of ellagic acid, the raw materials, which are the chestnut shells, have a cost 0.4 €/kg since it corresponds to 20% of the weight of a chestnut (Braga 2014). It also needs to be taken into account the price of a scoop of 25 g, 0.024 €/unit, and the packaging, 0.016 €/unit, which adds up to a total of 1.04 €/package.

Finally, considering a gross margin of 5 €, the selling price of each package of 500 g of pancake powder would be 6.04 €. It is noted that the extracted ellagic acid that was not used for the final product could also be sold as a powder at a very high value.

6. Conclusions
In this work, it was proposed a new product, Do Nut Stress – Acid ellagic enriched pancakes for anxiety control, consists of a pancake powder, considered a healthy and nutritious meal. This product intends to use ellagic acid extracted from chestnut shell. It was considered two stages: first, the ellagic acid extraction from the raw material and second, the production of the pancake powder. According to the literature, ellagic acid has anxiolytic properties, and it can be potentially used in consumers’ daily lives, preventing this type of illness. The pancake powder must contain a minimum of 0.03% of ellagic acid. However, this value should be
confirmed carry out experimental tests. It proposed a pack of 500 g of the pancake powder for the final product with a unitary price of 6.04 €.

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