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Formulation of Herbal Candies from Citrus Fruit Peels Powder and Spices

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Abstract

Citrus is one of the important varieties of fruits which include lemon, orange, sweet lime, kinnow, etc. This variety consumed by population in all over world by various processes. It is used by pharma industries and in research sector for making various products. The research content includes formation of candies from the combination of citrus fruit peels powder and spices. It is important for citrus fruits peels utilization. This research review have three types of candies formulated with different combination of peels powder and spices such as first supplement is formulated by

organic sweet lime peels with cardamom, second supplement is formulated by organic orange fruit peels with cumin seed and third is supplement formulated by organic lemon peel powder with combination of aniseed. Such kinds of product are impact able on health for human life to reduce the risk of gastrointestinal track problems and blood purification with normal activity of body. People should include such type of product in their routine to improve their lifestyle and to reduce the risk of disease.

Keywords: Citrus, Utilization, Formulation, Herbal

Introduction

Citrus is a universal term for plants belonging to family Rutaceae (Ladaniya, 2008) which considered as an important fruit around world and one-third of the crop is processed (Jiang *et al.*, 2014). This family has rich phytochemicals sources of many bioactive compounds which are responsible for antioxidant and many other biological activities (Fejzić and Čavar, 2014).

Citrus byproducts are promising sources of bioactive ingredients and of valuable technological and nutritional properties. These byproducts can be used as ingredients and food additives (Marín *et al.*, 2002; Puupponen-Pimia *et al.*, 2002; O'Shea *et al.*, 2012) in food industry for their cheap valuable component (Galanakis (2012). Peels are generated as the primary citrus byproducts represent about 50-65% of fruit weight during processing. These byproducts discarded and considered as a huge load to the environment (Mandalari *et al.*, 2006; Nayak *et al.*, 2015; Wang *et al.*, 2008; Ramful *et al.*, 2011).

Orange and lemon peels are common byproducts (wastes) produce from processing food and juice extraction industry. Lemon peels were applied for pectin and flavonoids (narirutin) production. Orange peels were also employed for recovery of flavonoids e.g., hesperidin, essential oils, and carotenoids. In Egypt and many Mediterranean countries, a major quantity of the citrus peels does not process. Some efforts were made to use these residues as livestock feed (Ghasemi *et al.*, 2009; Kim *et al.*, 2004; Masmoudi *et al.*, 2008; Chedea *et al.*, 2010^[11]; Di Mauro *et al.*, 1999^[14]; Farhat *et al.*, 2011; Bampidis, and Robinson, 2006^[5]).

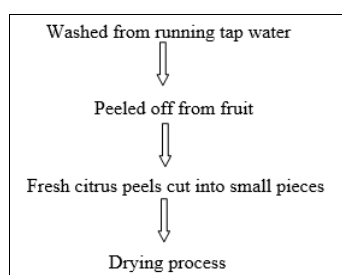
Natural products present in citrus peels e.g., sugars, flavonoids, carotenoids, folic acid, vitamin C, pectin and essential oils present are very useful for food industry and human health. Also, citrus peels are good source of phenolic compounds can be extracted and employed as natural antioxidants to prevent oxidation of some foods or may be utilized in designing functional foods (Patil *et al.*, 2009; Albishi *et al.*, 2013^[2]). Citrus peels described as rich source of unique phenolic compounds to citrus, especially the characteristic flavanone glycosides (mainly naringin, hesperidin, narirutin, and neo hesperidin). Huge amounts of flavanones and many polymethoxylated flavones which are very rare in other plants are contained in citrus peels (Bocco *et al.* 1998^[6]; Swapna and Bhaskar 2013). The antioxidant character of phenolics is due to their ability to donate an electron or hydrogen from phenolic hydroxyl groups. Phenoxy radical resultant tends to be poorly reactive because of electron delocalization in the aromatic ring, and therefore reactive radical is replaced by other one of limited activity (Li *et al.*, 2006; Shahidi and Naczak 2004; Topčagić 2009). β -carotene is a strongly red-orange pigment found in orange peels also their phenolic content has their contributions for quality attributes with color, bitterness, antioxidant and flavor (Delia-Gabriela Dumbravă *et al.* 2010^[13]; Kumar *et al.*, 2014; Legua *et al.*, 2014). β -carotene showed a nature strong antioxidant for avoiding and treatment of many diseases (Cooper *et al.*, 1999)^[12].

Antioxidants are a heterogeneous category of molecules which can safely interact with free radicals and stop the chain reaction before are damaged. Antioxidant capacity of food can use as an indicator of the beneficial effects on human health (Prior and Wu 2013). Antioxidants e.g., flavonoids, phenolic acids, vitamin C, vitamin E and tannins have different biological properties, such as anti-carcinogenic, anti-atherosclerotic effects, reduce coronary diseases and contribute to the maintenance of the gut health by modulation of microbial balance and these properties improve the quality and value of food & anti-aging (Lucia *et al.*, 2008; Kondo *et al.*, 2002; Tuberoso *et al.*, 2013; Liu 2004; Cai *et al.*, 2004^[9]; Ke *et al.*, 2015).

Antioxidant property is connected with the ability of phenolic compounds to scavenge free radicals, break radical chain reactions and chelate metals (Nayak *et al.*, 2015). The total antioxidant capacity of plant extracts is influenced by their chemical composition and antioxidant content. Antioxidants are greatly used as food additives to support degradation of foods and to improve their shelf life by preventing lipid per-oxidation as well protect oxidative damage (Kumaran and Karunakaran 2006). Therefore, natural antioxidants are needed for use in foods or medicinal materials and replace synthetic derivatives (Ramesh *et al.*, 2011). The antioxidant activity gives the ability of a bioactive compound to maintain cell structure and function by effectively clearing free radicals, inhibiting lipid peroxidation reactions and preventing other oxidative damage (Bravo, 1998)^[7]. Accordingly, citrus peels have been studied because they contain numerous biologically active compounds including natural antioxidants compounds (Hayat *et al.*, 2009). The antioxidant activity of orange flesh and peel extract containing compounds with different polarities, up to the knowledge, has not been reported. In addition, antioxidants may respond to different radical or oxidant sources in a different manner. Consequently, no single assay can accurately reflect all of the radical sources and antioxidants present in a mixed or complex system due to multiple reaction characteristics, mechanisms, and phase localizations which are usually involved (Prior *et al.*, 2005) Since the citrus fruit peels contain many valuable substances (natural products and bioactive phenolic compounds) can be changed into raw materials for intermediate food ingredients or as ingredients for value-added new products with various health benefits. Also, peels are considered as natural byproducts that can work as an outstanding low-cost antioxidant source.

Methodology

- Selection of citrus fruits** -: purchasing of organic lemon, sweet lime, orange from organic fruit store of Sonipat city.
- Processing of fruits** -: processing of fruits is classified in various steps -:



3. Drying Methods

Each of fresh sweet lime, lemon or orange peel pieces was divided separately into three parts and each part was dried using the following three methods:

a. Sun - Drying

Orange peel pieces were dried in sun light for 60 hr

b. Air Oven-Drying

Lemon peels pieces were dried in an air oven (Shellab-Model 1350FX.-Made in USA) at $40 \pm 2^\circ\text{C}$ for ~ 48 h.

c. Microwave -Drying

A programmable domestic microwave oven (type Samsung, 77 QH 400148, MF 2015, with a maximum output of 1500W at 2450 MHz) was used for drying the fresh sweet lime peel pieces samples for 6 min.

4. Formation of different peel powder

The three (sweet lime, lemon or orange) dried citrus peels were ground to a fine powder using a mechanical laboratory grinder and passed through a 24-mesh sieve, then package polyethylene bags and stored.

5. Formation of different spices powder

Different spices were used for formulation of spices powder

6. Formation of candies from different citrus peel powder and spices powder

Candy 1 -: Sweelofi -: sweelofi candy is formulated from the combination of sweet lime and cardamom.

Candy 2 -: lemofi -: lemoofi candy is formulated from the combination of lemon and aniseed.

Candy 3 -: Oranglofi -: orangofi candy is formulated from the combination of orange and cumin seed.

Analytical Methods

Proximate Chemical Composition

Moisture, ash, protein fat (ether extract) and crude fiber contents were determined in accordance with standard AOAC methods (AOAC 2005). Each analysis was carried out in triplicate.

Determination of Vitamin C content

The 2, 6-dichloroindophenol titrimetric method (Ramful *et al.*, 2010) was used to determine the vitamin C content of citrus peel extract. The tested peel sample(s) was blended with metaphosphoric acid -acetic acid solutions. After filtration and dilution, the diluted solutions were titrated against standard indophenols solutions. Results are expressed in mg ascorbic acid/g dry weight.

Determination of Total Phenolics Content

The Folin – Ciocalteu assay, adapted from (Singleton and Rossi 1965) was used for the determination of total phenolics present in the citrus peel extracts. Distilled water (3.5 mL) was added to 0.25 mL of diluted extract, followed by 0.25 mL of Folin – Ciocalteu reagent. A blank was prepared using 0.25 mL of 80% methanol instead of citrus peel extract. After 3 min, 1 mL of 20% sodium carbonate was added. Tube contents were vortexed then incubated for 40 min in a water-bath set at 40°C . The absorbance of the blue coloration formed was read at 685 nm against the blank standard. Total phenolics were calculated with respect to

gallic acid standard curve (concentration range: 0–12 $\mu\text{g mL}^{-1}$). Results were expressed in μg of gallic acid g^{-1} fresh weight of plant material.

Determination of Total Flavonoids Content

Colorimetric aluminum chloride method was used for flavonoids determination according to the methods described by (Ebrahimzadeh *et al.*, 2008) with some modifications. 0.5 ml solution of each sample extract was separately mixed with 1.5 ml methanol, 0.1 ml of 10% aluminum chloride, 0.1 ml of 1 M potassium acetate and 2.8 ml distilled water then left at room temperature for 30 min. The absorbance of the reaction mixture was measured at 415 nm with a double beam Perkin Elmer UV/Visible Spectrophotometer. Total flavonoid contents were calculated as quercetin from a calibration curve, which prepared by preparing quercetin solutions at concentrations 12.5 to 100 mg mL^{-1} in methanol.

Statistical Analysis

All the measurements were performed in triplicate and the data are presented as mean \pm SD. The obtained data were subjected to analysis of variance (ANOVA) according to PC-STAT, Version I A Copyright 1985, the university of Georgia, USA.

Result and Discussion Chemical Analyses

Proximate Chemical Composition

Proximate composition provides a general nutritional value of a food and includes analyses of the moisture, ash, protein, lipid content and crude fiber. Data in Table 1 showed the moisture contents of fresh lemon and orange peels & their samples dried by air oven (hot air) or microwave methods.

Fresh lemon peel sample contained more moisture content (81.23%) than orange sample (74.35%). Fresh citrus peel of Thompson navel, mandarin, and lemon are characterized by high moisture contents as reported by Nesrine *et al.*, (2012). After drying, the air oven lemon and orange peels still have more moisture than microwave dried samples without significant difference in between. As regard to microwave drying, lemon peels showed significant high moisture content compared to orange peels by $\sim 10.24\%$.

These findings agreed Adewole *et al.*, (2014)^[1].

This content revealed also %s of crude protein, fat (ether extract), fiber, and ash of control and both dried peel samples. Fresh lemon peels contained 11.53% crude protein whereas; orange peels had less crude protein by about 38.51%. Also, ether extract of dried lemon peel exhibited higher amount by 15.90% compared with dried orange peels. Total fiber contents of fresh lemon sample were greater than orange peels, i.e., 16.15 vs. 11.48 respectively. After drying, lemon peels still have more crude protein, total fiber and ash. With respect to ether extract, orange peels had significantly more amounts, reached to 2.44, 2.12% compared to 1.42 and 1.35% in lemon peels dried by the used two methods respectively. Regarding ash content, lemon peels had significantly more %s (5.92, 5.71) compared to (3.51 and 3.33) orange peels dried by microwave or air oven methods, respectively. These results agreed with Janati *et al.*, (2012) and Marfán, *et al.*, (2007) for total fiber, ether extract and protein. Also, Lemon peels had higher ashes content than orange (Nesrine *et al.*, 2012).

Ascorbic Acid Content

Vitamin C (L-ascorbic acid or simply ascorbate) is a water-soluble material. It is major in citrus and rich in the flesh & peel of fruits. It can efficiently scavenge diversity of reactive oxygen species (ROS), as a natural free radical scavenger, and give off semi dehydroascorbic acid, clearing $^1\text{O}_2$ and reducing sulfur radicals (Amitava and Kimberly, 2014)^[3].

Ascorbic acid contents of the investigated lemon and orange peel samples were determined by the 2, 6-dichloroindophenol titrimetric method. Drying of these citrus peels, either by microwave or air oven greatly reduced the ascorbic acid concentration to \sim less half content of their original values (control). Vitamin C loss in orange peels was of lower % than lemon peels after drying.

For example, its concentration in orange peels is reduced by ~ 47.89 and 47.93% after microwave and air oven drying, respectively. There is no significant difference in ascorbic acid content after drying by the two methods.

A similar trend was observed in lemon peels, although their loss was more than that happened in orange peels. The ascorbic acid loss in lemon peels, as a result of drying were 55.33 and 53.34% when drying was carried out by microwave or hot air (40°C), respectively. No significant difference was observed in ascorbic acid content as a result of the used drying methods. These results agreed with Fernández-Lopez *et al.*, (2004).

Total Phenolic Content

Polyphenolic compounds (as phenolic acids and flavonoids) are important fruit phytochemicals compounds for their antioxidant activities, their chelation of redox-active metal ions, and inactivation of lipid free radical chains and prevention of hydro peroxide conversion into reactive oxyradicals (Cabral de Oliveira *et al.*, 2009)^[8]. Phenolic content can be used as an indicator of antioxidant capacity and as a preliminary screen for any product when planned to utilize as a natural source of antioxidants in functional foods (Viuda-Martos *et al.*, 2011).

Data in Table 3 & Fig 1 illustrated that total phenolics (TPC) amount varied greatly and ranged in fresh to orange peel dried samples extracted with ethanol or methanol from 5255.02 \pm 24.04 to 1410.73 \pm 5.91 mg Gallic acid/ 100gm sample dry weight. The total phenolics content of orange peel extracted with ethanol was significantly higher ($p < 0.05$) than in methanol extract. No significant differences ($p > 0.05$) were observed in the phenolic levels of the two dried orange peels extracted with methanol. An opposite pattern was observed in dried orange peels extracted with ethanol compared to control samples. Meanwhile, ethanol extract exhibited higher phenolic content than lemon peel extracted with methanol and dried by microwave. On the contrary, a significant difference was found between TPC contents of air-dried lemon peel and microwave dried samples. Additionally, a presence of significant differences in the TPC content was noticed between all lemon peels extracted with ethanol. The noticed differences in the TPC may be related to nature and characteristics of the varieties of citrus fruit. The differences in the values of TPCS for various citrus peels types may be affected by environmental conditions, the degree of fruit ripening and genetic factors (Ladaniya, 2008). TPC of fresh peels are higher than the

recovery from dried samples because the water in fresh plant cells can help phenols extraction.

The reduction of phenolic compounds recovered from dried peels may be due to water evaporation and components in the cells (e.g., membranes and organelles) may hold together in the water absence and probably the extraction with solvent become more difficult. Moreover, if the citrus peel is dried before extraction, the recovery is much lower than using the fresh materials (Li *et al.*, 2006). The increase in drying temperature lead to a decrease in total polyphenols content after re-dissolution (Karshveva *et al.*, 2013).

Total Flavonoids Content

Citrus peels are rich source of natural flavonoids. Also, phenolic and flavonoid compounds of citrus have high antioxidant activity. Flavonoids possess a broad spectrum of chemical and biological activities including radical scavenging properties. Such properties are especially evident for flavonols (Kamran, *et al.*, 2009; Hayat *et al.*, 2010; El-Seedi *et al.*, 2012).

Total flavonoids content (TFC) of the investigated citrus peels is revealed in Table 4 Generally, TFC of the tested peel samples, extracted with methanol, were higher than those extracted with ethanol. Contents of the fresh orange peel samples, extracted with methanol, was 506.82 ± 0.97 ; meanwhile, the orange peel samples dried by microwave or air oven reduced to 309 ± 0.32 and 365.40 ± 0.16 QE/100g (db) respectively.

TFC content of dried peels with air oven was higher than microwave dried samples. Also, the same trend was noticed in the case of ethanolic extract. These findings varied from methanol extracts results for orange peel either fresh or dried samples which contained more (TFC) than ethanol extracts (Hegazy and Ibrahim 2012).

With regard to TFC content, data in Table 4 indicated that the lemon peel dried by air oven and extracted with methanol had the highest ($P < 0.05$) content (469.08 ± 0.42 mg quercetin equivalent / 100g db), followed by microwave dried and control samples with values 442.79 ± 0.42 and 430.58 ± 0.77 mg/100g db respectively. It was noticed also that TFC in the methanolic extract of lemon peels was higher than its corresponding ethanolic extract. Considering flavonoids, lemon gave the highest concentrations, which is agreed with reports available in literature.

Antioxidant Properties

Host of antioxidant phytochemicals found in citrus. Polyphenolic compounds (phenolic acids and flavonoids) are mainly responsible for fruits antioxidant activity. Total antioxidant capacity of food was measured using numerous methods. Different antioxidants may work through different mechanisms according to generation of different radicals and/or target molecules vary in their chemistry and in the way end points of these assays were measured. No single method can evaluate total antioxidant activity of foods. Two or more methods should always be employed to evaluate the total antioxidative effects of vegetables (Pellegrini, *et al.*, 2003; Nuutila *et al.*, 2003) The antioxidant activity of peel extract might be due to the reduction of superoxide anion, inactivation of free radicals, or complexation with metal ions or their combination (Karoui and Marzouk, 2013). Two complementary test systems: carotene– linoleic acid and DPPH were applied by Moulehi *et al.*, (2012) for evaluating the antioxidant capacities.

In the current work, the antioxidant activity of lemon and orange peels extracted with ethanol or methanol was evaluated using a range of antioxidant tests, including the Radical scavenging activities (DPPH), Trolox equivalent antiradical capacity (ABTS) and β -carotene.

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