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Research Article

Evaluation of antioxidant activity of watermelon peels (*Citrullus lanatus* L) and their application in cake making

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A B S T R A C T

Watermelon peels (WP) were treated by ethanol 70% (v/v) to extract flavonoids and phenolic acids then were identified by paper chromatographic technique. Synthetic antioxidant (butylated hydroxytoluene, BHT) and natural antioxidant (WPE) exhibited strong and close antioxidative activities (90.60% and 84.76%, respectively). The watermelon peels extract and BHT were added to sunflower oil at levels 100 and 200 ppm to keep its quality during heating at 180 ± 5°C for 28h. Moreover, the synthetic and natural antioxidants were added to cake made up by sunflower oil at the same levels. Cake was stored at refrigerator at 5°C for 28 days and the lipids were extracted every four days. The results reported that the addition of watermelon peels extract as natural antioxidant to sunflower oil and cake delayed the lipid peroxidation during heating oil and storage of cake.

INTRODUCTION

Antioxidants have an important and effective role in preserving food, especially foods rich in fats It has been known for a long time that dietary fats are exposed to oxidation thanks to their content of marital bonds that are present in the fatty acids involved in the composition of fats, due to many factors, which lead to the production of free radical and thus leads to the exposure of its consumer to many diseases, and the more fatty acids, the greater the marital ties and the greater the exposure of fats to oxidation and the production of an abundant amount of free radical that Lead to various diseases It was necessary to search for chemicals that act as synthetic antioxidants, for example dietary administration of butylated hydroxytoluene (BHT) , but after a period of use of the chemicals, it was found that they also have damage to health, such as kidney stress. Therefore, researchers began to search for natural compounds that play the role of antioxidant antioxidant activity, which is found in plants, algae, microorganisms and fungi, and the degree of safety is high, and therefore the search for these compounds continues, as they show their presence in a large proportion also in plant residues (crusts) and therefore these residues can be exploited in terms of obtaining natural compounds and also ridding the environment of pollutants that harm it, whether chemical or biological damage

Many extracts have been made on many plant residues to determine the quality of the extract and also the quality of the plant and show that most plants contain these compounds, but their percentage varies from one extract to another, although most of the extracts are or an aqueous

extract or an alcoholic extract. review - lemons - citrus fruits - watermelons - pumpkins ..... etc) such compounds (Allen and Hamilton, 1993 and Sulaiman *et al.*, 2013). Over time, free radical damage can cause problems with tissues and organs within the body, which can lead to disease. Research suggests that free radical molecules may join the risk of age-related health problems. Examples of these problems include heart disease, age-related macular degeneration, Alzheimer's disease and cancer. Your body's cells can repair or prevent damage caused by free radicals, for example by making antioxidant enzymes. The body gets certain antioxidants, such as vitamin C, from foods and drinks (Nanditha and Prabhasankar, 2009 and Sisein, 2014).

Koocheki *et al.*, 2007,watermelon is the fruit of plants of the family «cucurbitaceae», which is originally scientifically classified as vegetables, but in the majority of people watermelon is classified among the fruits due to the sweet taste of the pulp layer in it. Spherical or cylindrical watermelon fruit consists of 91 percent water, 6 percent of sugars, and the rest a wide variety of minerals, vitamins, fiber and more than a hundred chemical compounds with biofunctional effects in the human body. And each 100 grams of fresh watermelon pieces provides the body with only 30 calories. The body's need for vitamin C is provided by ten per cent, the body's need for vitamins A, B-1, B-2, B-3, B-5 and B-6 is provided by 4 per cent, and the minerals calcium, iron, manganese, magnesium, phosphorus, potassium and zinc by 3 per cent. But the most important advantage in the nutritional value of watermelon is the chemical compounds with biological effects in the body, such as carotenoids and others. Therefore, eating watermelon is mainly to provide fluids to the body from pieces of fruit that taste sweet and rich in bioactive chemical compounds in the body, especially lycopene compounds (antioxidant carotenoids) in the pulp and citrulline compounds (amino acids) in the pulp and skin. Perkins-Veazie and Collins, 2004 and Al-Sayed and Abdelrahman, 2013, explained that the chemical composition of watermelon contains many important compounds such as good source of vitamins, as well as a good source of lycopene (act as anti-oxidant during normal metabolism and protect against cancer). (Mandel *et al.*, 2005 and Muhammad, 2014) Hartati and Endah, (2015). The accumulation of free radicals in the body causes serious damage, and the high level of free radicals in the body is associated with many diseases, such as: cancer, diabetes, heart disease, arthritis, respiratory diseases, and immunodeficiency. The accumulation of free radicals in the body may also result in so-called oxidative stress, which causes cell damage and malfunction in various body functions.

There are two types of antioxidants. The body produces antioxidants known as autologous antioxidants, and there are external antioxidants obtained from natural plant or industrial sources. Exogenous antioxidants can be obtained from the following sources (Kumar, 1985 and Anthony, 2015).

The aim of this study was to evaluate some chemical properties of watermelon peels (WP) and separate and identify flavonoid and phenolic compounds in watermelon peels extracts. Add watermelon peel extract to cake to reduce oxidative stress and compare it with industrial antioxidants.

MATERIALS AND METHODS

Materials

Watermelon fruits were obtained from a hyper-supermarket in Zagazig city - Sharkia – Egypt. Peels were obtained after separating from full melon and cut into very small pieces and then dry in a vacuum oven at 40°C and ground into fine powder using laboratory electric mill then passed through 40 mesh sieves to separate the bran then stored in plastic bags in the freezer at -20°C until the extraction was camed out. Wheat flour, 72% extraction was obtained from Eastern Delta Flour Mills Company, Sharkia, Egypt. Ethanol and Butylated hydroxytoluene (BHT) was obtained from El-Nasr Pharmaceutical Chemical, El-Ameriea, Cairo, Egypt. Sunflower oil was obtained from Maser Company for Oils and Soaps, Zagazig Egypt. Sugar, egg, vanilla and backing powder were obtained from a hyper-supermarket in Zagazig City. And all reagent chemicals were of analytical grade and purchased from El-Gomhoria Co. Egypt.

Methods

Extraction and isolation of polyphenolic compounds from watermelon peels (WP).

20_mg of watermelon peel powder and melon peel samples were extracted in an ultrasonic water bath within **15** minutes using 750ml 70% methanol (phosphoric acid PH4). These samples were placed in a centrifuge for five minutes at a rate of 6000 rpm, then the floating samples were collected and the remaining samples were re-extracted using a larger amount of (500 ml 70% methanol). A brown product was obtained upon evaporation of ethanol of dryness and kept for flavonoid and phenolic compounds investigation according to Khoddami *et al.* (2013). The brown extract of watermelon peels were tested by paper chromatographic technique in order to identify the major flavonoid and phenolic compounds as described by Medić-Šarić *et al.* (2004). The watermelon peels extract and authentic samples were spotted on one dimensional Whatman No. 1 paper chromatography. The eluting solvents were butanol : acetic acid : water (4:1:5) and acetic acid 15% (ACOH). The different spots (major flavonoid and phenolic compounds and authentic samples) were located by color reaction and R_f value under UV lights with and without the presence of NH_3 fumes were identified according to Khoddami *et al.* (2013).

Preparation of watermelon peels extracts (WPE)

Watermelon peels extracts were prepared according to the method Mary, (2006) with some modifications 100g of The watermelon peel was separated from the washed fresh fruits, cut into small pieces, placed in trays to wash them with water, then dried at **50 C** using an air oven, and then placed in a laboratory to grind them and turn them into a fine powder.. The ethanol extract was concentrated using Soxhlet apparatus and the concentrated extract was used for analysis.

Chemical composition of watermelon peels

The degree of moisture, ash, protein, fat and dry fiber present in watermelon peel powders and melon peel as reported by the Official Analytical Chemists Association (AOAC) in 2000 was determined while the percentage of carbohydrates

was determined by degree of variation. - Absorbability of water and oilThe ability of both watermelon peel powder and melon peel to absorb water and oil is determined as reported by researcher in Dubois *et al.* (1956).

Determination of antioxidant activity

Flavonoid and phenolic compounds were evaluated as antioxidant activity of the previous extract from watermelon peels (WP) and compared with butylated hydroxytoluene (BHT) by thiocyanate method as described by Tsuda *et al.* (1993).

Addition of antioxidant to sunflower oil

Sunflower oil (commonly used for cooking in Egypt) was used as a substrate for oxidations studies. Natural antioxidant extracted from watermelon peels and synthetic antioxidant (BHT) were added to oil at 100 and 200 ppm on a dry weight basis to test their antioxidant effectiveness according to Buford, (1988). Sunflower oil with and without antioxidant (natural or synthetic) was heated in 500 ml glass beaker (Pyrex) at 180±5°C for 28 h (total heating hours) intermittent heating period was 4 h/day. The oil samples after heating were taken periodically and stored in glass bottles at -10°C till analysis.

Preparation of cake making

The ingredients of oil cakes making are given in Table (1) according to Hafez, (2012) with the modified method developed by Pinyon and Bamford in 1973. The recipe for the cakes is summarized in Table I, which shows the different proportions of watermelon peel powder and melon peel used in this study. To prepare, 'sugar and butter were put together and mixed for three minutes to become like cream' Then add the eggs to the mixture and beat with it for two minutes. After that, 'Add the sifted flour, baking powder and dried milk and beat the mixture for four minutes' After placing the mixture in a mold, stir for another minute The product was baked at 190°C for 25 min in an electric oven and cake was stored in refrigerator at 5°C and packaged in polyethylene bags for 28 days.

Table 1. Ingredients of cake made using sunflower oil

Ingredients	Flour	Sugar	Whole egg	Vanilla	Baking powder	Water	Sunflower oil
Weight (g)	100	125	75	0.5	6.5	20	50

Extraction of oil from cake making

Oil was extracted from cake samples every four days by soaking in n- hexane at room temperature for 48 h. The extract was filtered and evaporated to dryness. The extracted oils were kept in the deep freezer for further investigations.

Physico-chemical characteristics of oil

The weight and quantity of cake samples were determined one hour after baking (Randy Gill *et al*. 1995), and the weight-to-quantity ratio was calculated to obtain this specific amount. according to AOAC, (2000).

Determination of color of cake samples

Determination of the color of cake samples The color of cakes containing watermelon peel powder and melon peel was determined using spectra by means of a three-push color determination system. Color codes were also converted from x, y and z to l, a and b to conform to the formula developed by the manufacturers (Afshari-Jouybari and Farahnaky, (2011). Chromium (C), which represents the purity and purity of color, has been calculated from this equation:

C = A2 + B2 The total color density = (A2 + B2 + L2)1/2

Peroxide value of cake lipids

The lipids of cake were extracted using n-hexane as a solvent. Peroxide values were determined according to AOAC, (2000).

Sensory evaluation of cake samples

Ten panelists from the staff members of Food Science Department, Faculty of Agriculture, Zagazig University were asked to score the quality attributes of each cake sample and Determination of the amount of rotten cake samples Damage to cakes containing watermelon peel powder and melon peel was measured by testing the retention of alkaline water. Appearance, crust color, crumb color, crumb texture, taste, odor and overall acceptability were judged on a scale of 10 according to AACC, (1996).

Statistical analysis

Statistical analyses were conducted using SPSS version 15.0 for Windows. All analyses were performed in triplicate and data reported as means ± standard deviation (SD). Data were subjected statistically analyzed according to Steel and Torrie, (1980).

RESULTS AND DISCUSSION

Chemical composition of watermelon peels

The proximate chemical composition of watermelon peels under investigation was determined and the results are shown in the Table (2), it is clear that moisture content of watermelon peels was 11.42% while the total crude protein was 12.17%. Lipid content of watermelon peels was 3.94% but its crude fiber and total soluble carbohydrate contents were 16.58 and 55.12 %, respectively, and ash content of watermelon peels was 12.19%. it could be concluded that watermelon peels had total soluble sugars with the high value also crude proteins and ash with the moderate values, but it contained total lipids in low value. These data are in agreement with that obtained by Al-Sayed and Abdelrahman, (2013) and Muhammad, (2014) who reported that the chemical composition of watermelon peels showed nutrient value of fat, ash, protein, carbohydrate and fiber in dry weight (DW) as the values were 2.44, 13.21, 11.34, 58.73and 17.69 (g/100g), respectively.

Table 2. Chemical composition (%) of watermelon peels mg/100g

Moisture	Crude proteins	Total lipids	Crude fiber	Total soluble carbohydrates	Ash
11.42	12.17	3.94	16.58	55.12	12.19

Paper chromatography of watermelon peels extract

Flavonoids and phenolic compounds in WPE were identified with paper chromatographic technique and compared to authentic samples. Two solvents systems were used (BAW 4:1:5) and ACOH (15%) color reaction and R_f values of the flavonoids and phenolic acids compounds are shown in given Table (3). The ethanolic extract was found to contain five flavonoid compounds (Kampferol, quercetin, catechin, narginine and rutin) and three phenolic acid compounds (*P*. coumaric, ferulic and chlorogenic acids). These results are in agreement with those found by Oseni and Okoye, (2013) and Okafor *et al.* (2015) who found that the content of phenolic compounds in watermelon peels were characterized as flavonoids (Kaempferol, quercetin, catechin, naringin and rutin) and phenolic acids (*P*. coumaric, ferulic and chlorogenic acids).

Table 3. Polyphenolic compounds of watermelon peels extract

Compounds	Rf value		Without NH_3 fumes	With NH_3 fumes
	ACOH 15%	BAW	UV	UV
		Flavonoids		
Kaempferol	1	82	Yellow	Yellow
Quercetin	4	63	Yellow	Bright yellow
Catechin	67	85	Dark brown	Yellow
naringin	3	86	Deep purple	Greenish purple
Rutin	55	43	Deep purple	Yellow
		Phenolic acids		
***P*-coumaric**	91	67	Faint	Violet
Ferulic acid	52	72	Blue-violet	Green
Chlorogenic acid	63	63	Blue	Green

ACOH: acetic acid, UV: ultra violet light, BAW: butanol: acetic acid: water.

Antioxidant activity of watermelon peels extract

The efficiency of natural antioxidant of WPE was compared with synthetic antioxidant (butylated hydroxyl toluene, BHT) measured by thiocyanate method and the results are reported in Table (4). The natural and synthetic antioxidants showed strong antioxidant activity 84.42% and 90.22%, respectively. Lipid oxidation deterioration was the most cause of vegetable oils spoilage. Lipid oxidation do not only lower quality and nutritional value of foods, but also is associated with aging, membrane damage, heart diseases and cancer (Cosgrove *et al.*, 1987).

Table 4. Antioxidant activity of watermelon peels extract compared with BHT

	Absorbance at 500 nm	% Lipid peroxidation	Activity (%)
No additive	0.67	0.94	0.00
BHT*	0.045	7.12	90.22
Watermelon peels extract	0.090	13.40	84.42

*BHT: butylated hydroxytoluene

The peroxide value of oils extracted from sunflower cake was determined every four days up to twenty eight days and the results are given in Figure (1).

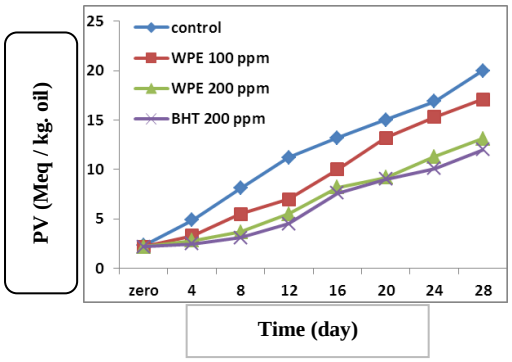

Figure 1. Peroxide value of sunflower oil extracted from cake after baking as affected with watermelon peels and BHT.

From the results, it could be noticed that 200 ppm of the watermelon peels extract and BHT effectively inhibited the peroxide formation for a period of four days (P.V 2.2 to 2.6 and 2.2 to 2.4 meq/kg). Then the peroxide value increased to 15.0 in natural antioxidants and 12.0 meq/kg at the end of store period. Very close results were observed for the addition of natural and BHT at 200 ppm. It is worth to mention that 200 ppm from watermelon peels extract also decreased the peroxidase value. Suggested that the addition of natural antioxidant from watermelon peels extract at 200 ppm delayed the peroxide value. On the basis the aforementioned results, it could be suggested that addition of natural antioxidant from watermelon peels extract at 200 ppm delayed the peroxide value.

Hunter-lab color values of cake samples

All color data are expressed as Hunter L*, a*, and b* values corresponding to lightness, redness, and yellowness, respectively. Hunter-lab color values of cake contained at different levels of WPE are given in Figure (2). Crust and crumb color of cakes varied with the quantity and the kind of the supplemented materials the crust became lighter (higher L*) as the watermelon peel extract level increased compared to that of control cake. No considerable differences in crust yellowness were found among the different WPE samples, both gave higher b* values than the control cake sample. Concerning crumb color, in generally, as WPE level increased L* and b* values decreased while a* value increased and the crumb color became darker and more greenish. The crumb of the control cake was lighter and more yellow compared to the tested cakes. It was well

known that during cake baking, the crumb, dose not reach a degree crumb does not reach degree above 100°C, so the Millard reactions fail to take place. Therefore, the resulted of crumb color of tested cake was due to the color of the used WPE and their interactions. These results were agreement with (Hafez, 2012).

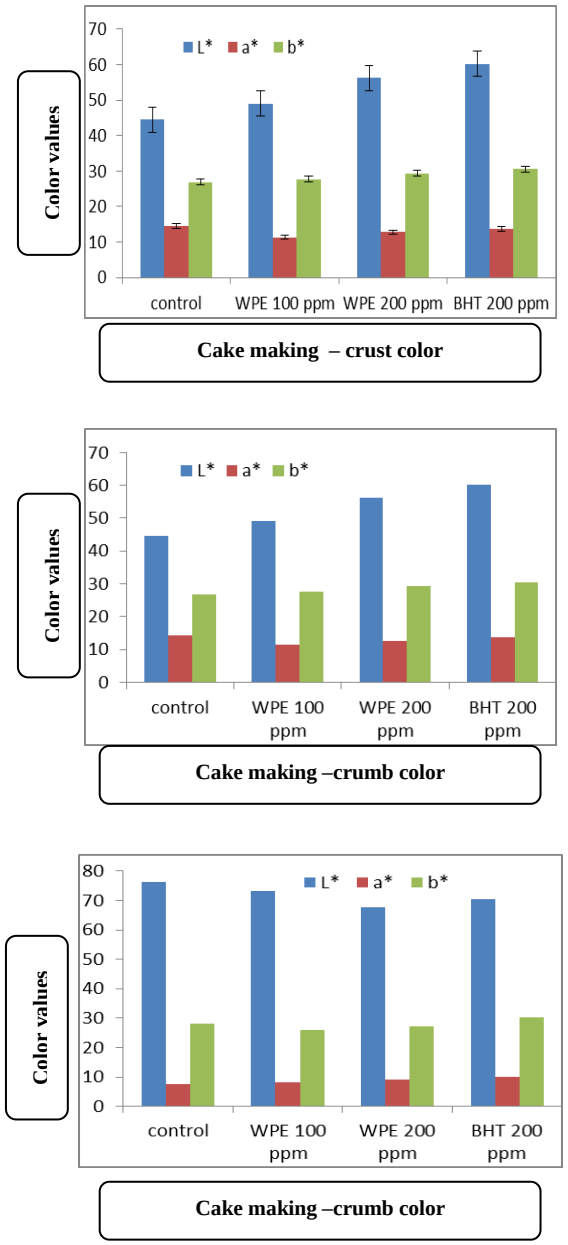

Figure 2. Hunter-lab color values of cake containing different levels of watermelon peels extract.

Sensory evaluation of cake samples during storage

The present data given in Table (5) showed sensory evaluation of cakes containing different levels of watermelon peels extract during storage at refrigerator at 5°C for 28 days. The sensory scores of cake containing different levels of watermelon peels extract were significantly affected. As the level of these materials increased, the received score values significantly decreased in all their quality attributes. Cake samples made from watermelon peels extract showed no significant differences in all their sensory properties and were as acceptable as those of control cake sample. Storage of different cake samples for 28 days significantly reduced all their sensory properties. The minimum reduction in sensory properties was observed in cake WPE followed while the maximum reduction was shows in control sample. These results revealed that WPE produced an acceptable cake which was not significantly different from the control cake. These results were agreement with those found by Al-Sayed and Abdelrahman, (2013) since they reported that there was no significant difference among the samples with and without cake preparation using watermelon peels powder substitution for the liking scores of appearances, color and odor exhibited.

Table 5. Sensory evaluation of cake prepared with watermelon peels extract.

Parameters	Control	WPE* 100 ppm	WPE 200 ppm	BHT* 200 ppm
Crump color	8.33 ± 1.09	7.00 ± 1.20	7.71 ± 1.65	6.81 ± 2.19
Crump texture	8.67 ± 0.63	7.12 ± 1.14	7.52 ± 0.89	7.86 ± 0.53
Taste	7.84 ± 1.04	6.62 ± 1.30	6.77 ± 0.74	6.84 ± 2.03
Odor	8.75 ± 1.10	6.62 ± 1.29	7.12 ± 1.06	8.03 ± 1.62
Over Acceptability	8.64 ± 0.75	6.97 ± 1.35	7.08 ± 1.22	6.71 ± 1.28

*WPE, watermelon peels extract, BHT, Butylated hydroxytoluene.

CONCLUSION

Natural antioxidants extracted from plant residues and their management are one of the latest major advances in food technology to solve the problems associated with their sustainable production, thereby improving their economic and health benefits due to their safer food applications and beneficial ingredients. Watermelon peel in vegetable production in Egypt and are consumed fresh or after processing processing, resulting in huge amounts of squamous peels and husks, as plant waste, that can be used as potential sources of antioxidants. Natural in the food industry. Watermelon peel extracts contain natural phytochemicals with superior protective effects that stabilize a variety of the most common vegetable oils consumed globally. Watermelon peels are good in antioxidant activity to increase shelf-life of cake and can be used in different industrial application

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