

EFFECT OF GAMMA RAY AND ELECTRON BEAM IRRADIATION ON THE PHYSICOCHEMICAL AND SENSORIAL PROPERTIES OF CHAMOMILE (*Matricaria chamomilla L.*)

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Abstract: The present study was carried out on dried chamomile powder to compare yield of extraction, physico-chemical characteristics and sensorial properties of extracts obtained from samples irradiated by gamma ray (GR) and electron beam (EB) at 10 kGy and 20 kGy. The results of the present study showed that radiation treatment with 10 kGy and 20 kGy of GR and EB did not affect significantly the yield of extraction. Also, the viscosity increased in the extract of samples treated with 20 kGy of GR and EB. However, both irradiation doses (10 kGy and 20 kGy) of GR and EB significantly increased the pH value, Mg concentration and L* value of color (lightness), and decrease the b* value of color (yellowness), but sensory qualities of all extract of the chamomile extracts samples were not significantly different depending on irradiation types and doses.

Key words: Chamomile, extraction yield, extract properties, electron beam, gamma ray, sensory evaluation.

Introduction

The actual tendency is to consider natural products as non-toxic and presenting few side effects than those used by conventional medicine (Petronilho *et al.*, 2012). Chamomile (*Matricaria chamomilla L.*) is a herb that has been in use since ancient times due to its many advantages and properties (Scalia *et al.*, 1999). Chamomile is one of the most important medicinal plants in the world trade that has many applications in drug and sanitary industrials (Petronilho *et al.*, 2012). Chamomile flowers are listed among the essential oils, and natural extractives that are generally recognize as safe (GRAS) for their intended use in food for human consumption (FDA, 2012).

Irradiation decontamination of herbs and spices is very popular in food irradiation because this process maintains their aromatic properties. This process has already been approved and is currently practiced (Kume *et al.*, 2009). Some chemical, physical and sensorial changes are induced in herbs as results of irradiation (Rico *et al.*, 2010; Shim *et al.*, 2009; Al-Bachir, 2007).

In fact, any treatment of herb subjected to, heating or ionizing radiation, there are changes in some of the herbs properties. Therefore, must be evaluated the effects caused by chemical and physical interactions of radiations with the irradiated product. The effect of irradiation, especially gamma ray, in chemical, physical and sensorial properties of different kinds

of herbs such as cowpea flowers (Abu *et al.*, 2006), nigella sativa seed (Khattak *et al.*, 2008), dehydrated paprika (Topuz and Ozdemir, 2004), medicinal herbs (Shim *et al.*, 2009), cinnamon (Salum *et al.*, 2009), red pepper (Rico *et al.*, 2010), lotus (Khattak *et al.*, 2009), turmeric (Dhanya *et al.*, 2009), tamarind seed (Choi *et al.*, 2009), licorice root powders and derivated products (Al-Bachir and Zeinou, 2005; Al-Bachir *et al.*, 2004; Al-Bachir and Lahham, 2003) and aniseed (Al-Bachir, 2007) are well known, but information about the effect of ionizing irradiation on the properties of chamomile is still very limited.

Therefore, the objective of this study was to investigate the effect of using gamma ray and electron beam irradiation on extraction yield and physical, sensorial and chemical properties of chamomile extracts.

Crude chamomile powder was irradiated with 10 kGy and 20 kGy by gamma rays (GR) and electron beam (EB), respectively and extraction yield (%), pH value, mineral concentration, viscosity and color changes of extracts were compared.

Materials and methods

Samples preparation and irradiation

Dried chamomile plant powder (4.5 kg) were purchased from the organic herb trading company in Damascus, Syria mixed and preserved in

Extraction yield

To obtain chamomile extracts from the commercially plant powder, 20 ml of distilled water were added to each sample of 5 g and mixed for 20 minutes. The suspension was made up to 200 ml with hot distilled water (75 °C) and mixed for 3 h (Al-Bachir and Lahham, 2003).

Total dissolved solids (dry weight-percentage) by concentrated extracts, obtained by drying for 6 h at 105 °C were determined in the extracts prepared immediately after irradiation according to the standard methods (AOAC, 2010).

polyethylene plastic bags (150 g per bags). They were divided into three groups: first group (6 bags) was non-irradiated control, the second group (12 bags; 6 bags for each dose) was irradiated at 10 kGy and 20 kGy of gamma ray (GR) with ⁶⁰Co source with a dose rate of 719 Gy hr⁻¹, at room temperature using gamma irradiator (ROBO, Russa). The source strength was approximately 55 kCi with dose rate of 712 Gy h⁻¹. The absorbed dose was assured by alcoholic chlorobenzene dosimeter. The absorbed dose was determined by the measurement of chloride ions or hydrogen ions by means Oscillotitrator (OK-302/2, Radelkisz, Budapest, Hungary), and the third group (12 bags; 6 bags for each dose) was irradiated at 10 kGy and 20 kGy of accelerated electron beam (EB). Irradiation was carried out using a linear electron accelerator facility (D-EPS-T30-30-002V, VIVARAD, France). The conditions of the accelerator were: beam energy 2.3 MeV (variable), beam power 2.6 kW. The beam current was 10 mA. The absorbed dose was measured employing film dosimetry (B3, GEX, corporation, USA). The absorbed dose was determined by measurement the optical density by means of spectrophotometer (Genesys 20, Thermo Fisher Scientific, USA). Irradiation was performed at room temperature and ambient pressure and the distance from the beam source to samples was 25 cm. One side irradiation (exposure from one sides) was performed. Chemical, physical and sensorial analyses were performed on the controls and treated sample extracts immediately after irradiation.

Chemical analysis of extracts

pH value of the chamomile concentrated extracts at was determined using an HI 8521 pH meter (Hanna Instruments, Woonsocket, RI, USA). Concentrations of Na⁺, K⁺, Mg₂⁺ and Ca₂⁺ cations were determined by adding lithium chloride to the solution using a Swiss flame photometer Model ATS 200 MKI (AOAC, 2010).

Physical analysis of extracts

To determine viscosity change, a suspension of commercially powder of chamomile was prepared according to methods of Hayashi (1996). The viscosity of the suspensions was measured with viscometer (HAAKE Viscotester 6 R plus, Thermo Electron, Kartsruhe, Germany) using a R2 column at

200 rpm. Viscosity values were determined and expressed as mPa.s. The color of chamomile powders extract was measured using AvaSpec Spectrometer Version (Avantes, Holland) at wave length of $\lambda=670$ nm by exposing the samples to the illuminant A light source and the CIE L* (lightness), a* (redness, and b* (yellowness) values were evaluated (Kwon *et al.*, 2009).

Sensory evaluation of the extracts

For the sensory evaluation, 25 ml of chamomile extracts were placed in a numerically coded weight glass beaker. A sensory test (using a consumer-type panel, comprised of 25 persons) was employed to detect sensory linking differences between treated and non-treated samples. Each member independently evaluated a chamomile extracts based texture, color, and aroma characteristics using a 5 point scale (1: extremely poor, 2: poor, 3: acceptable, 4: good 5: excellent) (Al-Bachir and Lahham, 2003).

Statistical analysis

The five treatments were distributed in a completely randomized design with three replicates. Data were subjected to the analysis of variance test (ANOVA)

using the SUPERANOVA computer package (Abacus Concepts Inc, Berkeley, CA, USA; 1998). A separation test on treatment means was conducted using Fisher's least significant differences (LSD) methods (Snedecor and Cochran, 1988) at 95% confidence level.

Results and discussion

Effect of gamma ray and electron beam irradiation on extraction yield

Extraction yields from chamomile powders in distilled water were determined and given in Table 1. The extract from non irradiated chamomile powder (control sample) gave an yield of 10.30 %. The yield of extraction was slightly decreased with increasing the dose of gamma ray (GR), while the yield of extraction was slightly increased with increasing the dose of electron beam (EB) radiation absorbed by the respective of solid samples. The effect of GR and EB beam irradiation on extraction yield of chamomile has not been investigation. However, earlier research studies showed different results for the effect of ionizing irradiation on the extraction dried weight yields of plant materials.

Table1. Effect of gamma ray and electron beam irradiation on total dry weight yield (%), electrical conductivity (EC) (ms/cm), acidity (pH value) and viscosity (mPa.s) of extracted from chamomile powder

Treatments	Extraction yield (%)	pH value	Viscosity (mPa.s)
Control	10.30±0.19	5.37±0.04	60.00±0.00
10 kGy-EB	10.77±0.22	4.96±0.04	60.00±0.00
20 kGy-EB	10.58±0.49	4.91±0.04	80.33±0.58
10 kGy-GR	10.06±0.18	5.09±0.04	60.00±0.00
20 kGy-GR	9.82±0.40	4.99±0.02	80.00±1.00
LSD 5%	0.58	0.07	0.94

EB: (Electron beam); GR: (Gamma irradiation)

Moreover, the current study indicated that, application of GR and EB did not affect significantly the yield of extracts of chamomile powders. Applying this assay, the same fining was previously found in extracts of several other herbs treated with

5 – 20 kGy gamma irradiation, e. g.: in licorice root powder (Al-Bachir and Lahham, 2003) or aniseed (Al-Bachir, 2007). On the other hand, gamma irradiation led to the increase of dried matter content and extraction yield of some kind of herbs with the

increase in radiation doses (Haung and Mau, 2007; Khattak *et al.*, 2009). In contrast, Polovka and Suhaj (2010) reported slight decrease in extraction yield of caraway irradiated with dose up to 30 kGy of gamma irradiation. The increase in the yield of extracts following irradiation might be due to degradation of some high molecular weight components, and changing these components from non-soluble to soluble ones in the test solvents (Harrison and Were, 2007). The difference in increase in extraction yield, as compared to that reported in the literatures may be due to different chemical composition of the plants (Khattak *et al.*, 2008).

Effect of gamma ray and electron beam irradiation on pH value

The results of the effect of gamma ray (GR) and electron beam (EB) on pH value of chamomile extracts are listed in Table 1. The pH value of the water suspension extracted from non-irradiated

control samples was 5.37. The pH value is one of the main factors influencing the quality of medicine. It always controls some chemical and microbiological reactions (Liu *et al.*, 2011). When the pH value is low (presence of acidic substances), the bacterial count could be low, but at neutral or higher pH the level of contamination of the herbal preparations could be observed to be higher. This suggests that a neutral or alkaline pH favored high microbial contamination levels of the herbal preparation. This agrees with observation that bacterial growth is optimal at more or less natural pH, around pH 5.0 – 8.5 (Abba *et al.*, 2009).

In our work regarding pH, statistically significant differences ($p < 0.05$) were determined between control group and irradiated samples at 10 kGy and 20 kGy of GR and EB. pH values of the groups non-irradiated and irradiated with 10 kGy (GR), 20 kGy (GR), 10 kGy (EB) and 20 kGy (EB) were 5.37, 4.96, 4.91, 5.09 and 4.99, respectively (Table 2).

Table 2. *Effect of gamma ray and electron beam irradiation on some minerals Mg, Ca Na and K (mg/Kg) content in extracts from chamomile powder*

Treatments	Mg (mg/Kg)	Ca (mg/Kg)	K/Na (mg/Kg)	K (mg/Kg)	Na (mg/Kg)	Ca/Mg (mg/Kg)
Control	8.02±0.96	7.54±0.91	3.00±0.005	11.31±1.36	3.77±0.45	0.96±0.24
10 kGy-EB	9.82±0.79	7.02±0.59	3.00±0.003	10.53±0.89	3.51±0.30	0.72±0.12
20 kGy-EB	10.49±0.56	7.57±0.67	3.00±0.003	11.36±1.00	3.79±0.33	0.73±0.10
10 kGy-GR	9.85±0.33	7.11 ±1.04	3.00±0.005	10.67±1.56	3.56±0.52	0.72±0.11
20 kGy-GR	12.94±1.71	6.55±1.60	3.00±0.004	9.83±2.40	3.28±0.80	0.50±0.06
LSD 5%	1.80	1.86	0.007	2.80	0.93	0.25

EB: (Electron beam); GR: (Gamma irradiation)

The decrease in pH value in irradiated chamomile powder sample was probably due to an increased amount of the organic acids released during irradiation treatments. Earlier research studies showed that irradiation treatment with 10 kGy did not significantly affect the pH value of the dried red pepper (Rico *et al.*, 2010). No significant changes in pH value of lyceum fruit were observed following 4, 8 and 15 kGy gamma irradiation (Wen *et al.*, 2006).

Similarly, pH value of four herbals, namely, rose, guggul, chirata and gulvel and four herbal formulations were not affected by gamma radiation treatment at the dose of 10 kGy (Kumar *et al.*, 2010). The content of polyphenolcarboxylic acids in chamomile decreased as the electron beam dose increased (Nemtanu *et al.*, 2008). Increased acidity of irradiated organic materials could be due to a breakdown of organic acids molecules, and it might

be due to the formation of carboxylic groups (Kim *et al.*, 2008). Sokhey and Chinnaswamy (1993) reported similar results, including that the breakdown of glycosidic linkages by the action of free radicals may cause an increase in the starch acidity.

Effect of gamma ray and electron beam irradiation on viscosity

The effects of gamma ray (GR) and electron beam (EB) irradiation on relative viscosity of extracts are shown in Table 1. It can be seen that, the viscosity of extract produced from non-irradiated control samples was (60 mPa.s), and irradiation with 10 kGy of GR and EB had no effect on viscosity, but irradiation with 20 kGy of GR and EB increased significantly the viscosity (80 mPa.s) of chamomile extract. In contrast, Farkas *et al.*, (1990) reported a dramatic decrease in the dispersion viscosity of heat gelatinized suspension of several irradiated spices with the starch content compared with that of un-irradiated samples.

The effect of irradiation seems to be related to the radio-depolymerisation of starch in irradiated spices (Gura *et al.*, 1997). The previous studies in our lab indicated that, immediately after irradiation at 5, 10, 15, and 20 kGy doses of gamma irradiation decreased significantly the viscosity of licorice extracts as compared with control (Al-Bachir and Lahham, 2003). The viscosity measurement was reported to be a promising method for the identification of irradiated herbs and spices (Farkas *et al.*, 1990).

Effect of gamma ray and electron beam irradiation on minerals contents

Adequate amount of potassium, calcium, magnesium and sodium were found in non-irradiated control sample of chamomile extracts (Table 2). Mineral components play an important role as adjuvant in the therapeutical activity of chamomile.

Wild chamomile plants are richer in distribution of mineral elements than the cultivated type, while ratios of K/Na and Ca/Mg in this cultivated type are many times higher than in wild chamomile (Maday

et al., 2000). The quantity of elements in any plant sample assumes importance, as it determines the nutritionally important minerals (Vadivel and Janardhanan, 2004).

Doses of 10 kGy and 20 kGy of Gamma ray and electron beam irradiation significantly ($p < 0.05$) increased magnesium in chamomile powder extract. The other three minerals studies (calcium, potassium and sodium) were not significantly affected by irradiation (Table 2). This is in disagreement with the earlier results reported by Al-Bachir and Lahham (2003); Al-Bachir and Zeinou, (2005); and Al-Bachir *et al.* (2004), who showed that 10 kGy of gamma irradiation increased the Ca, K and Na content in extract of licorice powders. Bhat and Sridhar (2008) found that electron beam irradiation significantly decreased potassium and calcium at 10 kGy onwards in lotus seeds.

Total potassium and sodium ratio ($K/Na=3.00$), and calcium and magnesium ratio of chamomile ($Ca/Mg=0.96$), are the quality index employed to determine the health benefits of the chamomile (Maday *et al.*, 2000). Also, the data suggests that there were no significant differences ($p > 0.05$) for the ratio K/Na (as defined in Table 2), between irradiated and non irradiated samples of chamomile extract. While 10 kGy and 20 kGy of gamma ray and electron beam irradiation decreased significantly the ratio of Ca/Mg of chamomile extracts.

Effect of gamma ray and electron beam irradiation on color

The color parameters of the extract of irradiated and non-irradiated chamomile powder, such as lightness (L^*), redness (a^*), yellowness (b^*) and color differences (E), were determined (Table 3). The extracts of irradiated chamomile powder became whiter and its degree of redness (a -value) and yellowness (b -value) was less intense.

The extract of irradiated samples constantly showed significant ($p < 0.05$) higher L^* value, and lower a^* value and b^* value than that of the control ones. In this study, the over all changes in color (E), of chamomile extract revealed that EB irradiation had a greater effect than the GR irradiation on the color changes of the chamomile extract.

Table 3. *Effect of gamma ray and electron beam irradiation on color of extracted chamomile powder*

Treatments	L**	a	b	E
Control	42.70±0.52	44.62±0.24	7.95±0.19	98.85±0.52
10 kGy-EB*	56.20±0.23	43.29±0.14	3.68±0.24	94.23±0.16
20 kGy-EB	54.70±0.35	45.54±0.08	3.96±0.43	95.77±0.27
10 kGy-GR	52.26±0.06	45.70±0.15	3.23±0.23	97.56±0.21
20 kGy-GR	45.94±0.24	44.38±0.20	5.45±0.70	98.64±0.36
LSD 5%	0.58	0.31	0.74	0.60

* EB: (Electron beam); GR: (Gamma irradiation)

**Color parameters: L, degree of whiteness (white +100 - 0 black); a, degree of redness (red +100 - -80 green); b, degree of yellowness (yellow +70 - 80 blue); and E, over all color difference

There is no information available in the literature on the effect of gamma ray (RG) and electron beam (EB) irradiation on the color of chamomile powder. However, for the other plant materials, diverse effects of irradiation on the color of its extract have been reported. The color changes of chamomile extract due to irradiation is in agreement with finding of Rico *et al.*, (2010) in dried red pepper, and Jeong *et al.*, (2009) in *Nelumbo nucifera*. Gamma irradiation slightly changed the color specification of the surface of the dried lyceum fruit samples: the lightness, redness, and yellowness of the samples slightly declined at high dosage (14 kGy) (Wen *et al.*, 2006). The results of the present study support previous studies showing that a leaf extract irradiated at a dose of 20 kGy and 50 kGy significantly increased the color L* value, while the

color b* values were decreased by irradiation. The differences in the average values of a* and b* between the turmeric samples due to irradiation was found to be statistically insignificant (p<0.05) (Dhanya *et al.*, 2009).

Effect of gamma ray and electron beam irradiation on sensory properties

The data on sensory evaluation (aroma, taste, color and texture) presented in Table 4, showed that all the samples (control, 10 kGy EB, 20 kGy EB, 10 kGy GR and 20 kGy GR) got higher scores for aroma, taste, color and texture with mean values around 4 from 5. Moreover, the extract of control samples showed maximum score of 4.19, 3.91 and 4.43, while the extract of 20 kGy GR showed minimum score of 3.91, 3.71 and 4.05 for aroma, taste, and color, respectively.

Table 4. *Effect of gamma ray and electron beam irradiation on the sensory properties of extracted chamomile powder (5 point scale).*

Treatments	Aroma	Taste	Color	Texture
Control	4.19±0.60	3.91±0.54	4.43±0.60	4.29±0.56
10 kGy-EB	4.19±0.75	3.95±0.67	4.14±0.73	4.38±0.59
20 kGy-EB	4.05±0.87	3.71±0.64	4.24±0.70	4.24±0.54
10 kGy-GR	4.10±0.77	3.76±0.63	4.24±0.54	4.19±0.60
20 kGy-GR	3.91±0.89	3.71±0.85	4.05±0.87	4.14±0.66
LSD 5%	0.48	0.41	0.43	0.36

EB: (Electron beam); GR: (Gamma irradiation)

The results are in good agreement with previously published papers on the influence of gamma irradiation treatment on licorice (Al-Bachir and Lahham, 2003, Al-Bachir and Zeinou, 2005; Al-Bachir *et al.*, 2004) and aniseed (Al-Bachir, 2007). As they found gamma irradiation of licorice powders and aniseed at dose of 10 kGy remained the sensorial properties of its extract unchanged. Similarly, triangle test panelists could not detect any significant difference between the 14 kGy gamma irradiated and non-irradiated lyceum fruit (Wen *et al.*, 2006). The scientific knowledge of chemical compounds is responsible for the characteristics odor and flavor of spices, which justify the food quality. Considerable efforts have been made to determine the irradiation effects on food components (Diehl, 2002).

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