Pharmaciana
Vol.9, No.1, May 2019, Page. 129-136
ISSN: 2088 4559; e-ISSN: 2477 0256
DOI: 10.12928/pharmaciana.v%vi%i.11893

Formulation of marigold (*Tagetes erecta* L.) flower extract in peel-off masks using polyvinyl alcohol and polyethylene glycol 6000 as the bases

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Submitted: 24-11-2018 ***Reviewed: 26-11-2018*** ***Accepted: 10-04-2019***

ABSTRACT

Flavonoids and carotenoids in marigold flower extract can act as a potent antioxidant. Because the use of antioxidant peel-off face masks is efficient, this research aimed to determine the effect of marigold (*Tagetes erecta* L.) extract, formulated with Polyvinyl Alcohol (PVA) and Polyethylene Glycol (PEG) 6000 as the bases, on the physical characteristics and stability of the produced peel-off face masks. This formulation used the ethanol extract of the marigold flower with three different concentrations, namely F1 3%, F2 4%, and F3 6%. The masks were then evaluated for their physical features (organoleptic, homogeneity, viscosity, spreadability, and drying time), chemical characteristic (pH), and stability. The results showed that all of the three formulas created a brown, soft textured, and homogeneous gel. Further tests revealed that F1, F2, and F3 differed in viscosity (18.466; 22.533; 28.133cps), drying time (29.1; 26.29; 24.64 minutes), pH (5.5I; 4.71; 4.59), and spreadability (6.7; 6.5; 5.9 cm) although not significantly. Based on both stability and freeze-thaw tests, these formulas were concluded as stable without any significant changes in their organoleptic properties and pH levels. Prepared with PVA and PEG 6000 as the bases, marigold extract can form peel-off face masks with good stability and physical features when used up to 6% concentration in the formulation.

Keywords: Marigold extract, peel-off mask, PVA, PEG 6000, antioxidant

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INTRODUCTION

Free radicals can cause premature aging of the skin. Skin aging can reduce skin elasticity and damage melanin (Lestari *et al.*, 2013). Naturally, the body has antioxidants known as enzymatic antioxidants, such as superoxide dismutase (SOD), catalase, glutathione peroxidase (GSH-PX), and glutathione reductase (GSH-R). However, when the skin is exposed to a significant number of free radicals coming from the environment, the antioxidants alone are not enough to ward off these free radicals. Therefore, antioxidants from outside sources are needed to keep the skin healthy (Winarsih, 2007).

One source of antioxidants is marigold flowers. These flowers are composed of two secondary metabolites, namely flavonoids and carotenoids, that have been proven to possess antioxidant properties (Gong *et al.*, 2012). Moreover, the ethanol extract of marigold flowers has a very strong antioxidant intensity, that is, IC_{50}= 7.6 ± 0.1 µg/mL (Valyova *et al.*, 2012). Another study has also reported that the IC_{50} of marigold flower extract is 3.4 µg/mL (Chivde et al., 2011). An antioxidant is considered very strong if it has IC_{50} <50 µg/mL (Phrutivorapongkul *et al.*, 2013).

Peel-off face masks are known for their unique characteristics because they use film-forming polymers that transform into a very cohesive plastic layer when dried completely, allowing for the manual removal of the product without leaving any residues. Moreover, these masks also provide a slight moisturizing action and enhance the effect of the active compound on the epithelium, especially as a result of the occlusive reaction exerted by the plastic layer.

In this study, marigold flower extract was formulated into peel-off face masks with three different concentrations and two bases, namely PVA and PEG 6000. This research aimed to determine the effects of varying levels of marigold flower extract on the physical characteristics and stability of the produced peel-off face masks.

MATERIALS AND METHOD

Materials

The marigold flower extract was obtained from Materia Medika Malang (No. 074/215A/102.7/2018).

Methods

The peel-off face mask was formulated with various concentrations of marigold flower extract (Table I). First, PVA was dispersed in 80 % of water heated at 80ºC (solution A) (Beringhs *et al.*, 2013). The dispersion was constantly homogenized until it was dissolved. The next step was dispersing PEG 6000 in water (solution B). Methylparaben and propylparaben were dissolved in propylene glycol and glycerin then mixed with the remaining amount of water (mixture C). Afterward, solution B was mixed with solution A until homogenous, then added with mixture C and stirred until homogenous. Finally, the marigold flower extract was poured into this mixture. Every formulation was left to rest for 48 hours before subjected to further evaluations to remove air incorporated in the formulation during the homogenization.

Table I. Formulation of marigold extract in peel-off face masks

Materials	Formula (% concentration)			
	Control	1	2	3
Marigold flower extract		3	4	6
Polyvinyl alcohol	15	15	15	15
PEG 6000	2	2	2	2
Propylene Glycol	10	10	10	10
Glycerin	5	5	5	5
Methylparaben	0.02	0.02	0.02	0.02
Propylparaben	0.18	0.18	0.18	0.18
Aqua Dest	ad 100	ad 100	ad 100	ad 100

Evaluation Methods

Organoleptic test

This test examined the smell, color, and shape of the formed gels.

Homogeneity test

A 0.1g sample was taken from the gels and placed on a glass object. This sample was then pressed with another glass object and observed to identify whether coarse grains still existed in the preparation (Tranggono and Latifah, 2007).

Viscosity test

This test used a Brookfield DV-E Viscometer, which can determine the resistance experienced by the spindle that has been dipped in thick material (Garg *et al.*, 2002).

Spreadability test

One gram of the preparation was weighed and placed between two glass plates. This procedure spread the test gels to all directions and created a diameter. Afterward, 135g weight was placed on the glass plate, and then the results were recorded (Zhelsiana *et al.*, 2016).

Drying time test

One gram of the preparation was weighed and applied evenly on the skin, covering an area of 7.5 cm x 7.5 cm. Then, the time needed by the mask to completely dry and form an easily peeled-off film layer was recorded as the drying time. Proper drying time is when the preparation has dried in less than 30 minutes (Vieira, 2009).

pH test

The pH was determined with a pH meter. One gram of the preparation was weighed and added with 10 mL of Aqua Dest. The pH meter was then calibrated. To obtain the pH levels, the electrode was rinsed with distilled water and dipped into the diluted preparation (Nibaho *et al.*, 2013).

Freeze-thaw test

The freeze-thaw method was carried out by storing the dosage forms at alternating temperature, namely 4ºC for two consecutive days and 40ºC for another two days (one cycle). The freeze-thaw test was carried out in four cycles (Mita *et al.*, 2015).

Stability test

The stability test was performed by weighing several samples of the preparation and putting them into dark vials. Each of the samples was stored in different environments, namely in storage with low temperature (5ºC ± 1.0ºC), in an oven (45ºC ± 2.0ºC), at room temperature with sun exposure (27ºC ± 2.0ºC), at room temperature and protected from sunlight (27ºC ± 2.0ºC). The characteristics and pH of these samples were analyzed every week for 28 days (Beringhs *et al.*, 2013).

Data Analysis

The data were processed statistically with analysis of variance (ANOVA) to identify their significance. A set of data was considered to be significant when the resulted coefficient was lower than 0.05 ($p<0.05$).

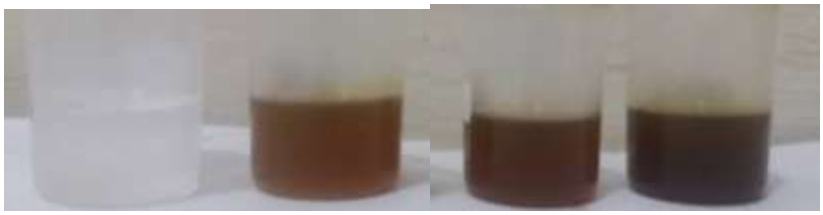
RESULTS AND DISCUSSION

The antioxidant properties of marigold flower extract

The marigold flower extract was tested for its antioxidant properties using the DPPH method, which can detect the anti-radicalization of test material accurately, reliably, and practically. Besides, this technique is simpler, faster, and more sensitive, and it only requires a few samples (Huang *et al.*, 2005). The results showed that the IC_{50} of the preparation was 57.77 ppm. An antioxidant is considered as very strong if it has IC_{50}<50 µg/mL (Phrutivorapongkul *et al.*, 2013).

Organoleptic evaluation results

The visible properties of the peel-off face masks prepared with different concentrations of marigold flower extract are presented in Figure 1. The solutions, labeled with Control, F1, F2, and F3, had different colors. The Control showed a clear color, whereas F1, F2, and F3 were yellowish brown due to the influence of the greenish-yellow marigold flower extract. All of the solutions had a soft texture and a distinctive odor. The homogeneity test aimed to determine the homogeneity of active ingredients and other additives in the preparations. The results showed that all of the test solutions had a homogeneous texture.



(A) (B) (C) (D)

Figure (1) Organoleptic evaluation and homogeneity test results of the marigold peel-off face masks. (A) Control or without marigold flower extract, (B) F1 was made with 3% marigold flower extract, (C) F2 with 4% marigold flower extract, and (D) F3 with 6% marigold flower extract

Viscosity

The viscosity test aimed to determine how thick the preparations were in this study and whether it influenced the dispersibility and release of the active substances. The results showed that, according to the SNI, the viscosity of the preparations was within the accepted range, namely 2,000-50,000 Cps (Table II). Increasing the concentration of the marigold flower extract produced a more viscous peel-

off face mask because of the lower water content in the formulation. The results of ANOVA showed that there was no significant difference between the viscosities of all preparations.

Spreadability

Table II shows that the spreadability of four preparations, i.e., Control, F1, F2, and F3, is within the range of good spreadability. The spreadability of consistent and comfortable-for-use semi solids varies between 5-7 cm (Fennema, 1996). The results of ANOVA revealed that the four preparations did not differ significantly in spreadability.

pH

The pH test aimed to determine the stability of the preparation and find out whether its administration would cause irritation when used on human skin. The pH levels of Control, F1, F2, and F3 were within the recommended range (to prevent irritation) and similar to the pH of the human skin, i.e., 4.5 - 8.0 (Table II). As the concentration of the marigold flower extract increased, the pH of the produced peel-off face masks lowered. The results of ANOVA revealed that the pH levels of all preparations were not significantly different.

Table II. The evaluation results of marigold flower extract in peel-off face masks

Formula	Viscosity (Cps)	Spreadability (cm)	pH	Drying time (minutes)
Control	16,800 ±1833	6.5 ± 0.2	6.03 ± 0.25	31 ±2.07
1	18,466 ±1747	6.7 ±0.3	5.50 ±0.11	29 ±0.43
2	22,533 ±5096	6.5 ± 0.25	4.71 ± 0.09	26 ± 0.27
3	28,133 ±3585	5.9 ± 0.25	4.59 ± 0.06	24 ± 0.39

Drying time

The drying time of the preparations was within 15-30 minutes, except for the control solution (Table II). The allowed drying time of a good semisolid is around 15-30 minutes (Vieira, 2009). The results of the drying time evaluation proved that increasing the concentration of marigold flower extract would reduce the drying time of the peel-off face masks because formulas with a high percentage of marigold extract had low water content. The results of ANOVA showed that there was no significant difference between the drying times of the four formulas.

Stability

The stability test aimed to determine whether the preparations could maintain the same characteristics as when they were prepared. These characteristics included identity, strength, quality, and purity throughout the specified shelf life (Joshita, 2008). During the four-week observation, the gel preparations did not show significant changes in organoleptic properties. They remained to have yellowish-brown color, distinctive scent, and soft texture. Subjected to different environments in the stability test—namely, stored at 5°C, 27°C without direct sun exposure, 27°C with direct sun exposure, and 45°C, the preparations showed increased pH levels (Table III). This condition is likely caused by the temperature-induced oxidation of the phenol compounds (Tranggono and Latifah, 2007) and degradation of carotenoid due to the heat (Wahyuni and Widjanarko, 2015). However, according to the SNI, the pH range after the stability test is still within the accepted threshold, that is, 4.5-8.0.

Table III. Comparison of pH levels before and after the stability and the freeze-thaw tests of the marigold peel-off face masks at various temperatures

Formula	pH					
	Initial	5 °C	27°C	27°C (sun)	45°C	Freeze-thaw
Control	6.03 ± 0.25	6.15 ± 0.23	6.12 ± 0.33	5.9 ± 0.14	7.17 ± 0.06	6.79 ± 0.38
1	5.50 ±0.11	5.26 ± 0.14	5.23 ± 0.14	5.08 ± 0.09	6.60 ± 0.14	5.54 ± 0.11
2	4.71 ± 0.09	5.22 ± 0.10	5.17 ± 0.08	5.07 ± 0.06	6.62 ± 0.09	5.34 ± 0.08
3	4.59 ± 0.06	5.10 ± 0.05	5.07 ± 0.07	4.96 ± 0.01	6.57 ± 0.13	5.20 ± 0.21

Freeze-thaw evaluation results

Instability occurs faster when dose preparations are stored in extreme conditions than in room temperature (Thanasukarn *et al.*, 2004). Based on the results of the four-cycle freeze-thaw evaluation, the organoleptic properties of the gel preparations did not change. The gels remained in yellowish-brown color, distinctive smell, and soft texture even after this test. However, there was an increase in pH levels. This condition is attributable to the oxidation of the phenol compounds induced by temperature difference (Tranggono and Latifah, 2007) and degradation of carotenoid due to the heat (Wahyuni and Widjanarko, 2015). However, according to the SNI, the pH levels after the freeze-thaw evaluation test are still within the allowed range, that is, 4.5-8.0 (Table III). The results of both the stability test and freeze-thaw evaluation showed that all formulas were stable and had no significant changes in their organoleptic properties and pH levels.

CONCLUSION

Marigold flower extract with concentrations of up to 6% can be formulated into peel-off face masks with PVA and PEG 6000 as the bases. All of the formulas produced preparations with good stability and good physical characteristics.

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