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Formulation and evaluation of herbal antioxidant face cream using extract of the marine seaweed *Sargassum spp*. (Phaeophyceae)

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ABSTRACT: The present study develops a formulation of face cream which is made by the ethanol extract of *Sargassum spp*. (Phaeophyceae) from the Jordanian Gulf of Aqaba. A different type of formulations (from F1 to F8), such as oil in water were discovered to obtain the best formula of a cosmetic cream of marine herbal origin. The evaluation of all formulations (F1 to F6) has been done by the analysis of different parameters like viscosity, spread ability and stability. An ethanol fraction analyzed from a sample of *Sargassum spp*. showed that F2, F3, F6 and F7 were of the best homogeneity. These formulas were the subject of further evaluation tests. By implementing the dye test to formulas F2, F3, F6 and F7, all were showed a O/W feature of emulsion. The evaluation of formulations of F3, F4, F6 and F7 was conducted by using an accelerated test. Parameters including homogeneity, appearance, type of smear and removal were tested and showed acceptable stability. Among the eight formulations (F1-F8), only F2 and F6 showed the best results in terms of stability, texture and evaluation test. F2 and F6 are held intact with good appearance and consistency that continues until day 15 after preparation. Hence, the study suggests that the composition of extract and the base of the cream F2 and F6 are more stable and safer.

KEYWORDS: Herbal cosmetics; emulsions; formulation; *Sargassum spp*.; phaeophyceae; Gulf of Aqaba.

1. INTRODUCTION

Human skin is the largest organ of human body, covering a surface area between 1.5 and 2.0 m^2. Therefore, human skin is considered the most accessible organ and it represents a promising route for drug delivery [1]. Since decades, transdermal route of administration is regarded an attractive delivery system among pharmaceutical researchers. Such route represents many advantages like avoiding first-pass metabolism by noninvasive means and can target the active ingredient for a local effect. People acceptance and compliance are another advantage whom they can easily and immediately apply topical products specially skin care cosmetics. Hence, achieving these aims could make topical products very interesting route and convenient for many users [2]. Therefore, cosmetics and skin care products are getting an increasing global interest by most of the pharmaceutical industries and researchers in recent years. The demand of new body and skin care products are mainly needed to serve different purposes like moisturizing, cleansing, exfoliating, anti-aging treatments and many more [3]. Indeed, herbal extracts incorporation in different kinds of cosmetics are showing promising results due to its contents of many natural compounds with healthy usage. Moreover, the natural contents in herbs specially of a marine origin have shown minimal side effects compared to compounds with synthetic origin [4,5]. High percentage of preservatives is incorporated in synthetic products which might result into allergic or irritation problems. Therefore, we need to use more pure natural cosmetics which are nourishing and contain rich supply of nutrients and minerals to support human skin and body [6]. Several substances that are found in many herbal extracts were reported to prevent oxidation in human skin and body, thus worked as good anti-oxidants [4,5]. Free radicals' production due to normal oxidation can cause damage to human skin cells. Indeed, increasing the number of free radicals could lead to wrinkling, photo aging, elastosis, drying and pigmentation of the skin [7]. Thus, the demand for anti-oxidant constituents

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of herbal origin is growing rapidly due to its curative activity against free radicals and reactive species, as well as its use as an anti-aging cosmetic [4,5]. Such substances can reduce the oxidative stress and terminate chain reactions by removing the free radical intermediates and inhibit other oxidation reactions by being oxidized themselves [7]. Marine organisms with thousands of species are found in all oceans worldwide and still many others are yet to be discovered [8,9]. This is beside the multiple discoveries and researches assigning their multitude of habitats in extreme environments. Many of these organisms are reported to produce different bioactive compounds [10,11]. Indeed, about 25,000 new biologically active compounds were identified [12]. Recently, bacteria and algae are getting an increased interest being the main producers of bioactive ingredients among other marine organisms. Marine seaweed represents a source of minerals, polysaccharides, proteins, lipids, and secondary metabolites such as phenolic compounds, terpenoïds, halogenated compounds as well as many others [13,14]. The three major categories of marine algae, namely, Chlorophyta (green algae), Rhodophyta (red algae) and Phaeophyta (brown algae) were reported of having compounds with varying bioactive substances that might have pharmaceutical applications [15]. Antioxidant activity is reported in numerous genera of marine algae, including *Ahnfeltiopsis*, *Colpomenia*, *Gracilaria*, *Halymenia*, *Hydroclathrus*, *Laurencia*, *Padina*, *Polysiphonia*, and *Turbinaria* [16]. Such antioxidants from marine algae are reported to play important role against various diseases and aging factors in human [17]. It further has potential anti-inflammatory, antibacterial, antifungal, anti-proliferative and anticancer properties [16,17]. *Sargassum spp*. is reported to contain high levels of substances with antioxidant activity [18,19]. Therefore, the purpose of this study was to develop a justified formulation of face cream which is made by the ethanol extract of *Sargassum spp*. (Phaeophyceae) that has suitable physical properties capable to provide the fairness and anti-aging effect in human skin from the Jordanian Gulf of Aqaba.

2. RESULTS

The results of accelerated stability test are shown in Table 1. Formulation F2 and F6 showed more stability in O/W type emulsion. F2 produce a uniform distribution of extracts in cream. When F3 and F7 were kept for a long time, it was found that there are changes in the color of the cream. After application of cream F2 and F6, the type of smear formed on the skin was not greasy. The cream F2 and F6 were easily removable by washing with water.

Table 1. Accelerated stability testing protocol

Days	Temperature	Formulations	Parameters			
			P_1	P_2	P_3	P_4
0	Room temperature (40±) °C	F2	***	NCC	NG	***
		F3	***	NCC	NG	***
		F6	***	NCC	NG	***
		F7	***	NCC	NG	***
5	Room temperature (40±) °C	F2	***	NCC	NG	***
		F3	*	CIC	G	*
		F6	**	NCC	NG	***
		F7	*	CIC	G	*
10	Room temperature (40±) °C	F2	***	NCC	NG	***
		F3	*	CIC	G	*
		F6	**	NCC	NG	***
		F7	*	CIC	G	*
15	Room temperature (40±) °C	F2	***	NCC	NG	***
		F3	*	CIC	G	*
		F6	**	NCC	NG	***
		F7	*	CIC	G	*
20	Room temperature (40±) °C	F2	***	NCC	NG	***
		F3	*	CIC	G	*
		F6	**	NCC	NG	***
		F7	*	CIC	G	*

P_1: Homogeneity; **P_2**: Appearance; **P_3**: Type of smear**; P_4**: Removal.

***: Good; **: Satisfactory; *: Bad. **NG**: Non greasy; **G**: greasy; **ES**: Easy; **NCC**: No change in color; **CIC**: change in color.

3. DISCUSSION

Marine brown seaweed is used as medicinal ingredients and healthy foods in Japan, China, and Korea [21]. Seaweed have attracted considerable interest as potential functional metabolites [22] and functional components in pharmaceutical, food, and cosmetic industries [23,24].

Seaweed has specific chemical constituents which contributed much in its bioactive properties. Recently, interests are significantly increasing by the use of natural remedies (plants extracts) in many of the artificial cosmetics [25,26]. Thus, availability of new herbal ingredients from seaweed, the financial rewards upon developing successful formulations as well as the consumer demand for safe products all are showing an upward trend in cosmetic industry. Seaweed is the major source of natural products that are used in cosmetology nowadays. Seaweed contains different biochemical compounds including polysaccharides, proteins, lipids, phenolic compounds, pigments, vitamins, and other bioactive substances [27]. Indeed, seaweed are able to produce both primary metabolites (i.e., polysaccharides, proteins, amino acids, fatty acids) as well as secondary metabolites including pigments, phenolic compounds, sterols, vitamins, and several other bioactive agents [28]. Moreover, seaweed was also reported to have a protective effect against damaged skin from UV radiation or simply against aging due to its antioxidant activity. The production of free radicals in the body causes oxidative stress and oxidative photo damage of macromolecules and plasma membrane components in the skin. Ingredients of seaweed possessing antioxidant activity that makes them very suitable in face or skin care cream or serums due to its anti-aging, anti-wrinkles, as well as sun protection effects. Hence extracts of seaweed were found of good choice of ingredient incorporation in many face creams. This is certainly with other ingredients incorporation in cream formulas (i.e., Almond oil) for skin's moisturizing and glowing [29].

Sargassum spp. has been widely used for folk medicine [30], and because of its potential, Sargassum spp. might be an excellent candidate for the cosmetic industry. Sargassum is a genus of brown seaweed of the Sargassaceae family which has been demonstrated to exhibit antimicrobial, antioxidant, anti-inflammatory, antitumor, antipyretic, analgesic, antiedema, neuroprotective, anticoagulative, and hepatoprotective activities [30]. Sargassum spp. are among the most dominant observed species of marine brown seaweed in the Jordanian Gulf of Aqaba [31]. The Jordanian Gulf of Aqaba is located at the east fork of the Red Sea with arid climate of high evaporation (~400 cm/year) and negligible precipitation (~2.2 cm/year) and runoff. Average water temperature is 23.5˚C and salinity is about 40.5. The biodiversity of the Gulf of Aqaba is rich and unique compared to other marine ecosystems of similar climate conditions [32].

The results of this study are beneficial for the effective use of Sargassum spp. extract as a justified formulation of face cream. Our study indicated that all the formulations of the prepared cream from the extract of Sargassum spp. were O/W type emulsion. Only the formulations (F2 and F6) were more stable, whilst remaining formulations were not stable when stored for long time. These formulations (F2, F3, F6 and F7) had showed an important information in O/W type emulsion about homogeneity, appearance, greasiness and color of the formula.

These formulations (F4 and F5) were fully separated into two immiscible layers typically upon increasing the extract percentage for up to more than 1%. Only F1 emulsions were not homogeneous and multiple lumps and aggregations were formed having a bad impact on emulsion type cream consistency. In addition, F8, was fully separated after immediate formulation. These formulations (F2, F3, F6 and F7) were proved to have the best homogeneity. Therefore, their formulas were selected and subjected for further cream evaluation and suitability tests. Dye test indicated that F2, F3, F6 and F7 are having a O/W feature of emulsion. Results on the test of accelerated stability are shown in Table 1. In samples examined from F3 and F7 were showed contamination after five days since the commencement of the accelerated study. From above discussion, it is concluded that the formulation F2 and F6 are held intact with good appearance and consistency that continues until day 15 after preparation. The research work suggests that the best formula for further studies it is highly recommended to be based using the parameters shown in Table 2.

Table 2. Composition of *Sargassum* spp. extract-based face cream (g).

Ingredients	F1	F2	F3	F4	F5	F6	F7	F8
Stearic acid	15	1	1	1	1	2	2	1
Cetyl alcohol	0.5	4	4	4	4	3	3	2
Almond oil	10	4	4	4	4	4	4	4
Paraffin oil	10	8	8	8	8	8	8	8
Propylene glycol	12	4	4	4	4	4	4	4
Triethanolamine	0.5	2	2	2	2	2	2	2
Glycerol	7	4	4	4	4	4	4	4
Methyl paraben	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03
Propyl paraben	0.015	0.07	0.07	0.07	0.07	0.07	0.07	0.07
Ethanol extract	1	0.5	1	2	3	0.5	1	1
Water	Qs	Qs	Qs	Qs	Qs	Qs	Qs	Qs

F: formula; **Qs**: Quantity sufficient.

4. CONCLUSION

F2 and F6 are held intact with good appearance and consistency that continues until day 15 after preparation. Hence, the study suggests that the composition of extract and the base of the cream F2 and F6 are more stable and safer.

5. MATERIALS AND METHODS

1.1. Seaweed sampling

The brown seaweed (*Sargassum ssp.)* samples were collected in the spring of 2020 from the beach of the Marine Science Station (MSS) in the Gulf of Aqaba (29.4582° N, 34.9767° E). *Sargassum ssp.* samples were dried at room temperature for several days, grinded into fine powder, and then placed inside a clean container and frozen at 4°C until extraction was performed.

1.2. Preparation of extracts

Shade-dried and coarsely powdered samples of *Sargassum spp*. (714.2g) was subjected to Soxhlet extractor for extraction using petroleum ether and then successively extracted with ethanol. The extracts were then concentrated to dryness under reduced pressure and controlled temperature ,and kept in refrigerator at 4°C for further use.

1.3. Cream formulation

An oil/water (O/W) emulsion-based cream (semisolid formulation) was formulated according to hydrophilicity of the extract and high solubility in water-based formula. The emulsifier (stearic acid) and other oil soluble components (acetyl alcohol, almond oil) were dissolved in the oil phase (Part A) and heated to 70°C. The preservatives and other water-soluble components (methyl paraben, propyl paraben, triethanolamine, propylene glycol and the ethanol extract of *Sargassum spp.*) were dissolved in the aqueous phase (Part B) and heated to 70°C. After heating, the aqueous phase was added in portions to the oil phase with continuous stirring. Perfume was added and the formula for the cream is given in Table 2.

1.4. Evaluation of cream

1.5. Type of emulsion under dye test

The scarlet red dye is mixed with the formulated (O/W) cream. A drop of the cream was placed on a microscopic slide and examined under microscope. If the dispersed globules appear red and the ground is colorless, the cream is O/W type. The reverse condition occurs in W/O type cream (i.e., the disperse globules appear colorless in the red ground) [20].

1.6. Accelerated stability testing

Accelerated stability testing was the second approach that was performed by studying the most accepted formulas. Four stable samples studied for 7 days at room temperature. The formulation numbers were F2, F3, F6 and F7 at room temperature and elevated temperature (40+1°C). The formulations were

observed and tested on 0th, 5th, 10th, 15th and 20th day for homogeneity, visual appearance, touching affinity, color, pearl essence and roughness (Table 1). The type of film or smear formed on the skin was then checked up immediately after the application of the cream. Finally, the ease of removal of the cream off the skin was examined by washing the applied part with tap water.

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