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Original Article

Application of factorial design in the development of cosmetic formulations with carrageenan and argan oil

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Abstract

Background: Gel-cream formulations used as cosmetics can present various textures. They may be fluid or semisolid, being highly influenced by their composition. These presentations require a complete study to obtain information about the physical stability and consistency of the product and the possible interaction between its compounds. In this context, this study aimed to develop cosmetic formulations using carrageenan and argan oil and assess through factorial design the influencing factors on the physical-mechanical properties.

Materials and Methods: This study presents two different moments, totalizing 28 formulations developed. First, 27 formulations were formulated according to a 3^3 factorial design study to understand the influences of concentrations, processes, and interactions between the raw material. In a second moment, four formulations were selected from this factorial: F1 (1% carrageenan, without argan oil); F2 (2% argan oil; without carrageenan); F3 (without both), and F4, the formulation with the presence of both in the highest concentration. The formulation F4 was utilized to create F5, which presents the same basic composition as F4, but added 0.1% of NaCl. All the formulations were studied regarding the texture parameters using the TA.XT Texturometer.

Results: The natural ingredients carrageenan and argan oil presented a synergistic action in improving the physical-mechanical properties of the formulation. It is possible to observe that the viscosity index is the most affected parameter according to the presence or absence of the variation of the raw materials. The addition of NaCl increased significatively all the physical-mechanical parameters studied. Thus, the formulation F4 presented higher results for consistency and index of viscosity when compared with F1, with the only carrageenan.

Conclusions: This work has a significant contribution once showed that the use of factorial design could contribute to developing more suitable cosmetic formulations, besides evidence the possible interactions between the raw materials from natural sources, giving subsidies for the development of more stable cosmetics and with superior performance.

Keywords: Factorial design, Argan oil, Cosmetics, Carrageenan, Texture

Background

Gel-cream formulations represent an essential strategy in the development of cosmetics. They can be characterized as emulsions stabilized with hydrophilic colloid, conferring applicability to receiving other ingredients such as active substances. This way, stable and effective topical formulations can be achieved in the development.[1]

An important global movement is the sustainability of practices, which aims to reduce impacts on the environment.[2] One of the strategies for sustainability used in the cosmetic industry is stabilizing the formulation using hydrocolloids, reducing the number of emulsifiers utilized in the system.[3] However, the utilization of acrylate polymers in cosmetic products has been contested since consumers are afraid of the impacts of these ingredients in nature.[4] On the other hand, it is challenging to utilize polymers from natural sources, which presents a competitive and/or superior performance compared with synthetics. Thus, the study of natural polymers and hydrocolloid ingredients applied to cosmetic formulations needs to be explored.

In this context, the utilization of carrageenan as a thickener agent in cosmetic formulations can be an alternative for synthetic polymers. However, few studies in the literature explore the utilization of this polysaccharide in the cosmetic field.[5]

Carrageenan is a group of natural polysaccharides present in the algal cell structure of the Rhodophyceae type.[5,6] They may form colloids and gels in aqueous at low concentrations, being this another advantage. These gels are transparent, achieving a wide variety of textures, from very elastic and cohesive, to firm and brittle formulations, depending on the combination of the polysaccharides fractions utilized in the formulation.[5]

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These polysaccharides can thicken in the presence of ions such as potassium and calcium.[6]

At the same time, in the development of a gel-cream formulation, the utilization of oils is significant since it can improve the skin barrier and parameters related to skin hydration.[7] Thus, argan oil can be an alternative in developing a topical formulation that can be applied in the cosmetic field.[8] argan oil is produced in the process of cold pressing kernels of argan tree seeds. Because of its rich range of ingredients like fatty acids such as unsaturated fatty acids, tocopherols, carotenoids, phenolic compounds, sterols and squalene.[9] The presence in it of tocopherols and polyphenols with their antioxidant effect neutralizes free radicals and protects the skin against harmful effect of sun light radiation.[8,9] Besides, it can improve skin parameters which may be compromised in the aging process.[10]

Gel-cream formulations used as cosmetics can present various textures. They may be fluid or semisolid, being highly influenced by its composition.[1] These presentations require a complete study to obtain information about the physical stability and consistency of the product and the possible interaction between its compounds.[11,12] Therefore, the choice of raw materials is the most important step in cosmetic research and development, significantly when cosmetics from natural sources are developed.[13] Waxes, polymers, surfactants, and other ingredients may significantly influence properties of formulations such as stability, efficacy, acceptability and sensory perception.[1]

The study regarding the interactions between raw materials from the natural origin in topical formulations is significant in cosmetology. However, these interactions can cause stability problems affecting stability, performance, and sensory, requiring a detailed investigation of the contribution of each raw material, as well as possible combinations and concentration influences. [11] One way of evaluating possible interactions is to1use statistical design of experiments.[2,11,12]

Calixto et al[11] investigated the correlation between waxes and polymers in the physical-mechanical characteristics of cosmetic products and the impacts in the sensorial perception using the design of experiments. Ferreira et al[12] utilized the factorial design to understand how emulsifiers and their concentrations can influence rheological parameters for sunscreen formulations. The utilization of factorial design according to the physical-mechanical characteristics, such as firmness and viscosity, can be a helpful tool to improve cosmetic development. However, this type of study needs to be more explored since its applications are vast and, sometimes, specific.[2] The utilization of a factorial design is a way to improve the cosmetic development assertiveness, saving money and time in the development.

In this context, the application factorial design contributes to optimizing natural cosmetics development through statistical analysis of the physical-mechanical evaluation impacting the final product. This research is fundamental to bring subsidies for further investigations and cosmetic development. In this context, this study aimed to develop cosmetic formulations using carrageenan and argan oil and assess through factorial design the influencing factors on the physical-mechanical properties.

Materials and Methods

Materials

The corn (Maizena®, Brazil) and tapioca (DaTerrinha®, Brazil) starches used in the present study were obtained at a local market (Ribeirao Preto, SP, Brazil). Vegetal Glycerin, butylene glycol, disodium EDTA, argan Oil, Cartagena Iota and phenoxyethanol, methylparaben, ethylparaben, propylparaben, butylparaben, and isobutylparaben were provided by Mapric (Sao Paulo, SP, Brazil). The emulsifier utilized was Polyglyceryl-6-Distearate (Gattefosé). The classical emulsion formulation was used to prepared the formulations.

Formulation Development

This study presents two different moments, totalizing 28 formulations developed. First, 27 formulations were formulated according to the factorial design study to understand the influences of concentrations, processes, and interactions between the raw material.

In a second moment, four formulations were selected from this factorial: F1 (1% carrageenan, without argan oil); F2 (2% argan Oil; without carrageenan); F3 (without both), and F4, the formulation with the presence of both in the highest concentration. The formulation F2 was utilized to create F5, which presents the same basic composition as F2, but added 0.1% of NaCl. This second part of the study aimed to understand the influence of sodium chloride in the presence of carrageenan.

Experimental Design

The design of experiments used a statistical strategy that optimizes experimental processes and rationalizes the decision regarding cosmetic development. It permits the simultaneous study of the effects of different variables on a response, in this case, how ingredients and processes can improve the stability of cosmetic products. [2,11,12] A general full factorial experimental design with three levels was elaborated for the copolymer and emollient under study.

Two continuous numeric factors were defined to assess the influence of the presence and concentration of the ingredients on the formulations, Carrageenan Iota concentration (0%, 0.5%, and 1%) and argan Oil concentration (0%, 1%, and 2%). Besides, the agitation velocity was defined as a possible variation (process variations), and the studied velocities were 800 rpm, 1200 rpm, and 1600 rpm, being a study 3.[3] The stability predictor variable "firmness" was chosen because this physical-mechanical property is a predictor of cosmetic products stability. The results were compared according to concentrations, concentrations and agitation, velocity, and the presence and/or combination of the ingredients. Twenty-seven formulations were developed in the total, and they were analyzed by TA.XT plus Texture Analyzer (Stable Microsystems, United Kingdom) equipped with the probe Back Extrusion rig A/BE 35 mm.

Texture Analysis

The texture analysis was performed using a TA.XT plus Texture Analyzer (Stable Microsystems, United Kingdom) equipped with the probe Back Extrusion ring A/BE 35 mm at room temperature. The method consists of the insertion of the analytical probe into the sample, with defined speed and depth, leading to a pre-defined period of recovery between the end of the first compression and the beginning of the second, resulting in a force (g) versus time (t) graph. This test evaluated the parameters: index of viscosity, consistency, firmness, and cohesiveness, being firmness utilized in the experimental design. For this, the formulations were loaded in 125mL containers, which are 50 mm in diameter. For the test, 75% of the container needs to be completed with the formulation. In this test, firmness is obtained from the maximum value of the positive curve. The return distance used was 25 mm, the return speed was 20 mm s^{-1}, and the minimum contact force was 30 g.[12] The formulations were tested 24 hours after their development.

Statistics

The experimental data obtained were submitted to statistical analysis. The Shapiro-Wilk test was used to evaluate the normality of the populations. A one-way analysis of variance with the Tukey post-test was used (α = 0.05). In the case of non-normal distribution, the Kruskal–Wallis with Dunn's post-test was applied (α = 0.05).

Results

Formulation Development and Factorial Design

To reduce the utilization of other oils, which could interfere with argan oil results and increase the formulation stability, we utilized a cosmetic base with starches developed by our research group.[4] The emulsifier utilized was polyglyceryl-6-distearate since of its high skin compatibility and natural origin. The formulation presented glycerin from natural origin and butylene glycol as humectants—the formulations present 96% of ingredients from natural sources.

Regarding the factorial design, it was possible to observe that the agitation velocity did not influence too much in the physical-mechanical parameters, being 800 rpm the most interesting for the increase in the texture parameters. The ingredients concentration influences linearly in the firmness parameters, as expected. However, the combination of carrageenan and argan oil showed a synergistic behavior for firmness, especially in high concentrations (Figure 1). The formulation with a higher concentration of argan oil and carrageenan iota and lower agitation speed (800 rpm) was selected to be added with 0.1% of sodium chloride.

Influence of the Ingredients and Sodium Chloride

The formulations F1, F2, F3, F4, and F5 were tested regarding the index of viscosity, consistency, firmness, and cohesiveness texture parameters. The results are presented in Figure 2, and the comparison according to each formulation using the Tukey post-test is presented in Table 1. It is possible to observe that the viscosity index is the most affected parameter according to the presence or absence of the raw materials. The addition of NaCl increased significatively all the physical-mechanical parameters studied. Thus, the formulation F4, developed with the factorial design as the one with the synergistic interaction, presented higher results for consistency and index of viscosity when compared with F1, with the only carrageenan.

Discussion

The present study brings relevant data regarding the development of phytocosmetic formulations based on carrageenan and argan oil. It was possible to observe a synergistic interaction between these ingredients in the firmness parameter using the factorial design.

The increase of the physical-mechanical characteristics in a synergistic combination can be correlated with the carrageenan proprieties. The algae polysaccharide increases the cosmetic formulation viscosity in the presence of ions, molecules with charge.[6] The utilization of oil in combination increased the physical-mechanical characteristics of the formulations, probably because lipids are amphiphilic molecules with charge density. This way, the increase in the texture parameters observed in a synergistic form can be related to the interaction between these two ingredients. This superior increase is not just related to the increase in the concentration as observed in Figure 1, but especially when both ingredients are together in the formulation, evidencing the interactions.

To study this theory, we developed the formulation F5 using the formulation obtained in the factorial design with the highest firmness value (F4) and adding 0.1% of NaCl. As observed in Figure 2, all the texture parameters increased after the salt addition. On the other hand, this increase was too extreme and can compromise the sensory properties and acceptance of the product,[11] evidencing that the addition of NaCl – even in low concentrations - cannot be desirable for topical formulations.

The results for F4 were more satisfactory, presenting values for texture parameters that may be more satisfactory in the sensory evaluation. Other studies already correlated the texture with the sensory, and it is observed that formulations with a good balance on these

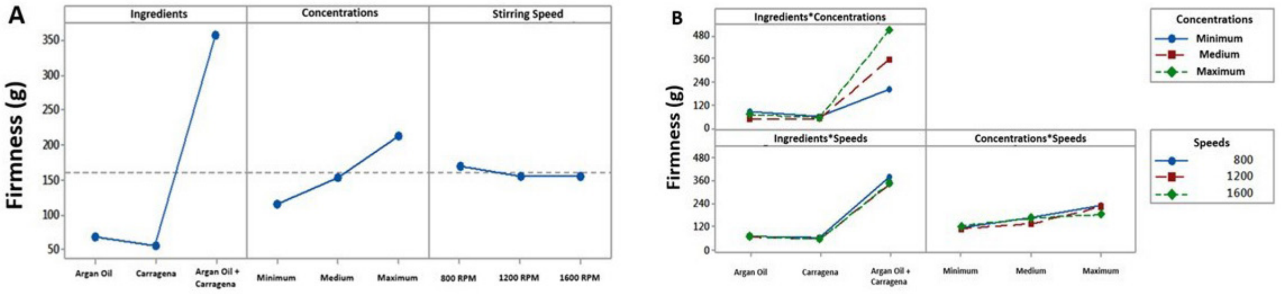

Figure 1. Factorial Design Results From the Interaction Between Carrageenan, Argan Oil, and Stirring Speed. The first graphics (A) represents the synergistic interaction between ingredients. The other graphics (B) showed that ingredients and concentrations are the most expressive for the firmness parameter.

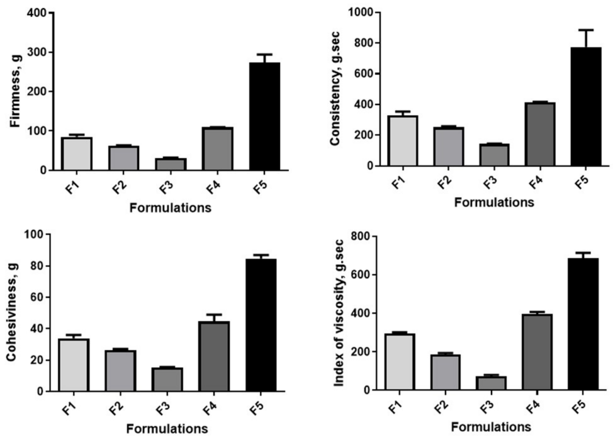

Figure 2. Results for the Physical-Mechanical Parameters Evaluated for the Formulations With the Presence, Absence, or Combination of Carrageenan and Argan Oil. The formulation F5 also presents NaCl on its composition, evidencing the thickening effect of the carrageenan.

Table 1. *P* Values According to the Tukey Post-test for Each Physical-Mechanical Parameter

Comparison	Firmness	Cohesiveness	Consistency	Index of Viscosity
F1 vs F2	ns	ns	0.02	<0.001
F1 vs F3	0.002	<0.001	<0.001	<0.001
F1 vs F4	ns	ns	0.004	<0.001
F1 vs F5	<0.001	<0.001	<0.001	<0.001
F2 vs F3	0.03	ns	0.003	<0.001
F2 vs F4	<0.001	0.03	<0.001	<0.001
F2 vs F5	<0.001	<0.001	<0.001	<0.001
F3 vs F4	<0.001	<0.001	<0.001	<0.001
F3 vs F5	<0.001	<0.001	<0.001	<0.001
F4 vs F5	<0.001	<0.001	<0.001	<0.001

Note: ns means not significant.

parameters may present better consumer acceptance and performance.[2,11,13-15]

The results observed for F4, when compared with F5, evidenced that the oil in combination increased the physical-mechanical characteristics of the formulation F4, probably because lipids are amphiphilic molecules, with charge density, however not so much as observed for F5, with the cation in its composition. This way, it is possible to increase the viscosity of semisolid formulations using carrageenan without use salt in the composition, not exceeding the values that can affect the perception and performance. This information is new in the literature and can be useful in future perspectives regarding the development of phytocosmetics.

According to the physical-mechanical results, the principal differences observed among the formulations F1, F2, F3, and F4 were correlated with the intrusion analysis. The Back Extrusion test presents a ring that is introduced and extruded from the formulation with controlled velocity.[11] In the intrusion moment is observed the firmness and the consistency. Firmness is the highest point in the curve, and consistency is the area under the intrusion curve. In the extrusion, the lowest point is characterized as cohesiveness, and the area under the extrusion curve is the viscosity index.[11,14] The synergistic interaction between carrageenan and argan oil is observed, especially in the extrusion moment (Figure 2). However, the firmness parameter is also affected, as observed in Figure 2, and it is not more effective than the presence of carrageenan without salt.

This information evidence that the formulation is benefited by the synergistic interaction, especially in the rearrangement after applied stress. This way, it is possible to observe that F4 is superior to F1, F2, and F3 because of the better interactions between the raw materials in the formulation and better plasticity once the stress did not affect the structure rearrangement.

Conclusions

The ingredients carrageenan and argan oil presented a synergistic action in the improvement of the physical-mechanical characteristics. These results can be related to the physical-chemical characteristics of the lipids, amphiphilic molecules, allowing the carrageenan thickener action.

Finally, this work has a significant contribution once showed that the use of factorial design could contribute to the development of more suitable cosmetic formulations, besides shreds of evidence the possible interactions between the raw materials from natural sources, giving subsidies for the development of more stable cosmetics and with superior performance.

Competing Interests

None.

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