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The Use of Silicones as Extractants of Biologically Active Substances from Vegetable Raw Materials

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Abstract: Based on theoretical studies, the authors of this paper propose the use of cosmetic organosilicon polymers (commonly called silicones) for the extraction of a complex of biologically active substances contained in vegetable raw materials. It is important to note that the biological molecules do not interact with the organosilicones and, therefore, their properties are not altered after the extraction. In this work, we investigate the efficiency of several polyorganosiloxanes as extractants of vegetable raw materials (*Calendula Officialis* L. and *Artemisia Absinthium* L.) useful for the preparation of cosmetic emulsions. Specifically, the extraction studies were conducted by using polyorganosiloxanes with a single component (polydimethylsiloxane Silicone Oil 350 cSt, cyclopentasiloxane BRB CM 50, and phenyltrimethicone BRB PTM 20) as well as a mixture (PEG-12 polydimethylsiloxane BRB 526, a solution of dimethiconol in cyclopentasiloxane BRB 1834, and amodimethicone BRB 1288). Compared to water and ethyl alcohol, polyorganosiloxanes are more effective in the extraction of the biologically active substances that are contained in the raw plants. Interestingly, the combination of different polyorganosiloxanes improved the extraction efficiency. The attained knowledge can be helpful in the development of a novel protocol for the formulation of emulsions appealing for cosmetic applications.

Keywords: extraction; cosmetic polyorganosiloxanes; biologically active substances; aqueous and silicone plant extracts; antioxidant activity

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1. Introduction

Vegetable raw materials have been largely employed in cosmetics due to the presence of a broad range of biologically active substances with different functionalities. Recently, numerous studies have been aimed towards the development of cosmetic formulations containing growing amounts of natural molecules [1–3]. Biologically active substances from plants belong to different classes of inorganic and organic compounds, which possess a specific activity that influences the catalytic and regulatory functions in the human body [4]. Remarkably, the properties of the biological molecules are affected by both the raw plant and the extraction procedure. Moreover, it should be evidenced that the functionalities of biological compounds can be controlled by their loading within inorganic nanomaterials [5–7].

It is well known that water represents the traditional extractant used within cosmetics. Nevertheless, it should be noted that aqueous solvents allow the extraction of only water soluble substances from the plants. According to several studies [8–10], the disadvantages of the extraction by water include the absence of essential oils, the low content of flavonoids, and the microbiological instability of extracts, which requires further heat and/or chemical treatments. Within the cosmetic industry, both ethyl alcohol and water–ethyl alcohol mixtures are common solvents employed for the extraction of flavonoids, coumarins, and cinnamic acids, and other polyphenolic compounds, saponins, terpenoids,

and chlorophylls. [11]. However, ethyl alcohol is a flammable substance and, consequently, fire safety requirements are needed during the extraction procedure. In addition, ethyl alcohol has a dehydrating effect, dries the skin, and its removal from the extract requires additional technological operations of evaporation and drying [12]. In principle, other solvents (hexane, diethyl ether, and acetone) could be used for the extraction of biologically active molecules from plant raw materials. Although they are efficient, they cannot be used for the production of cosmetics because of their toxicity towards humans.

In addition, the presence of the stage of purification of the extract from the solvent leads to an increase in the cost of production [13]. Therefore, the most promising protocols in the cosmetic formulations require the optimization of the extraction of the several biologically active substances in their native state by using different types of extractants. Literature reports that the optimization of the final composition of the cosmetic products can be achieved by using their ingredients (such as glycerin, propylene glycol, butylene glycol, capri/caprate glycerides, silicones, and vegetable oils) for the extraction, avoiding further removal steps [14].

It is well known that silicones possess interesting properties for cosmetic production [15,16]. They are odorless and they present low evaporation, heat high vapor density, and the relevant thermal and oxidative stability. Their viscosity is almost independent of temperature. Silicones are resistant to UV radiation and they are not an appropriate substance for the development of microflora, which contributes to the safety of cosmetics. As reported in a review [17], some polyorganosiloxanes can be bioaccumulative and harmful to human health.

In addition, silicones are chemically inert substances. Therefore, based on theoretical studies, we propose the use of cosmetic organosilicon polymers as extractants suitable for a complex of biologically active substances contained in the raw material. Remarkably, organosilicones do not interact with the biological molecules, which preserves their properties after the extraction [15,16,18]. In this study, we observed that the combination of polyorganosiloxanes as mixtures can be a perspective route to improve the extraction efficiency in vegetable raw materials, such as *Calendula Officialis* L. and *Artemisia Absinthium* L.

2. Materials and Methods

2.1. Materials

Silicones are from BRB International BV (Netherlands). Polydimethylsiloxane Silicone Oil 350 cSt; cyclopentasiloxane BRB CM 50; phenyltrimethicone BRB PTM 20; PEG-12 polydimethylsiloxane BRB 526; a solution of dimethiconol in cyclopentasiloxane BRB 1834; and amodimethicone BRB 1288 have been selected (Table 1).

Table 1. Chemical structure of polyorganosiloxanes.

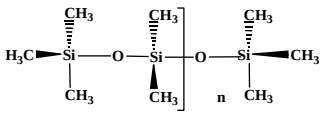
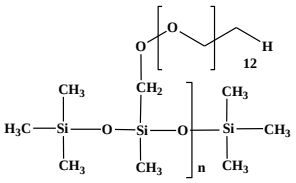
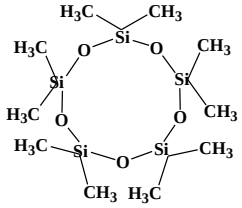
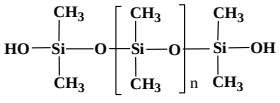
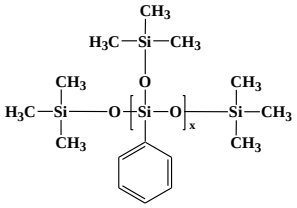
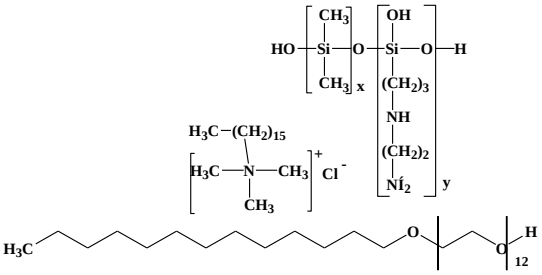
Chemical Structure	Chemical Structure
 Silicone Oil 350 cst	 BRB 526
 BRB CM 50	 BRB 1834

Table 1. *Cont.*

Chemical Structure	Chemical Structure
 BRB PTM 20	 BRB 1288

2.2. Methods

2.2.1. Characterization of Silicones

The determination of the refractive index of silicones was performed using the refractometric method [19]. To determine the refractive index of silicones, a refractometer type IRF-22 was used.

The density of silicones was determined using the pycnometric method [20].

The viscosity of silicones was determined using the Ostwald method [21]. The research method is based on measuring the duration of the liquid leakage time from the glass viscometer.

The determination of the surface tension of silicones was performed using the method of ring separation [22]. This method consists of determining the amount of force required to separate the ring wetted by the test liquid from its surface.

The marginal wetting angle of silicones was determined according to [23]. The method of determination is based on the measurement of the wetting angle, which is calculated using the height and diameter of the size of the droplets applied to the solid surface.

Relative spread studies were performed using a modified method of Zeidler U. [24] in vitro on dense gelatin plates. Test samples of silicones were applied (at least 6 drops weighing 4 mg) on prepared, dried gelatin plates. After 10 min, the oil impression was taken using filter paper and the area occupied by one drop was determined. The result was the arithmetic mean of the areas of all the drops for each silicone sample.

2.2.2. Extraction Procedure

Among the great variety of plant extracts, two groups can be distinguished: extracts from common plant species and exotic plant species (extracts obtained from very rare plants).

The common species of plants include any medicinal vegetation that is grown and collected in a certain area and is characteristic of the area, adapted to the peculiarities of the climatic conditions. In particular, the common plant raw materials typical of the territory of Ukraine were selected for research purposes, namely the dried calendula plant and wormwood. Domestically produced aquatic plant extracts from herbaceous plants (calendula (*Calendula Officialis* L.) and wormwood (*Artemisia Absinthium* L.)) were used.

Calendula (*Calendula Officialis* L.) and wormwood (*Artemisia Absinthium* L.) were tested for extraction. Plant matter was dried at a temperature of 25–30 °C in a well-ventilated room with a moisture content of up to 14%, which is optimal for the destruction of cell membranes and the extraction of their contents. The crushing of raw materials was carried out on roller crushers until the size of 3–5 mm was reached. The homogeneity was assessed by sieve analysis using a set of pharmacopoeial sieves. The mass ratio of raw materials and extractants was was fixed at 1:5. The prepared raw material was weighed and placed in a heat-resistant container with a ground lid. Then, the containers were filled with the calculated amount of extractant (selected polyorganosiloxane) and mixed. The resulting mixture was capped and placed in a thermostat for 24 h at the constant temperature of 40 °C. During the extraction, the mixture was periodically stirred. After the extraction

process, the extract was separated from the solid meal by filtration and defended (Figure 1). The final extract was used for further characterization.

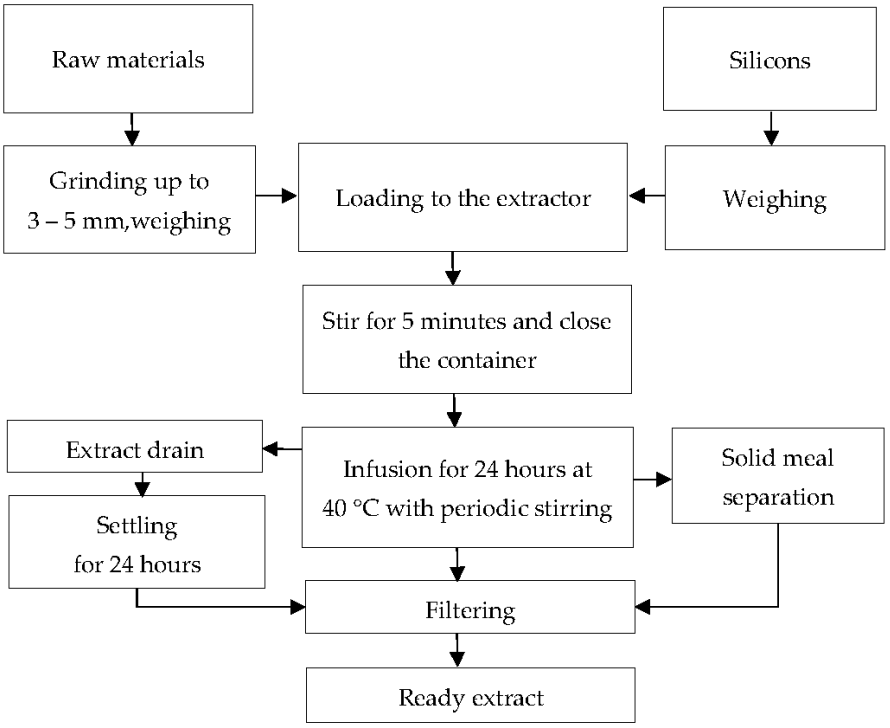


Figure 1. Technological scheme of vegetable raw material extraction silicone extractants.

2.2.3. Characterization of the Extracts

The vitamins (B_1, B_2, B_5, B_6, P, PP, C, A, and E) in the investigated extracts was determined by qualitative analysis based on the qualitative chemical reactions [25]. The quantitative content of vitamin C in plant extracts was determined using the iodometric method [26]. The method is based on the oxidation of the vitamin C molecule by iodate ions in an acidic environment.

The quantitative content of flavonoids in extracts was determined using spectrophotometric method. We selected 4 mL of the test extract (solution A), which was placed in a 25 mL volumetric flask, into which we added 2 mL of a 2% ethyl alcohol solution of aluminum chloride and brought the volume to the mark with 95% ethyl alcohol. After 20 min, we measured the optical density on a spectrophotometer at a wavelength of 410 nm in a cell with a layer thickness of 10 mm. For comparison, we prepared solution B as described below. We inserted 4 mL of solution A in a 25 mL volumetric flask into which we added 1–2 drops of 1% hydrochloric acid. Then, the solution volume was brough to the mark by adding 95% ethyl alcohol. The content of flavonoids in terms of avicularin (expressed as percent X_F in the dried raw material) was calculated as:

$$X_F = (D \cdot 25 \cdot 100)/(330 \cdot m), \quad (1)$$

where D is the optical density of the test extract; 25 is the total volume of the sample taken for determination; 330 is the specific absorption rate of avicularin complex with aluminum chloride at 410 nm; and m is the mass of the sample expressed in grams.

The content of tannins was determined using titrimetric methods. An amount of 2 mL of the final extract was selected and placed in a conical flask with a capacity of 100 mL. Then, we added 60 mL of distilled water and 2 mL of indigosulfonic acid solution (1 g of indigo carmine was dissolved in 25 mL of concentrated sulfuric acid, then an additional 25 mL of concentrated sulfuric acid was added and diluted with water to 1000 mL by carefully pouring the resulting solution into water in a volumetric flask with a capacity of

1000 mL, and then stirring it). The obtained mixture was titrated with 0.1 mol/L potassium permanganate solution to a golden yellow color. Separately, we conducted a control experiment by titrating 2 mL of indigosulfonic acid in 60 mL of distilled water with 1 mL of 0.1 mol/L potassium permanganate solution. This corresponds to 4.157 mg of tannins according to Equation (2) and takes into account the conversion factor reported in the literature [27]. The content of tannins is determined in percent X_T by the following formula:

$$X_T = ((V_1 - V_2) \cdot K \cdot D \cdot V \cdot 100)/m \cdot V_3, \quad (2)$$

where V_1 and V_2 are the volumes (mL) of the potassium permanganate solutions for the titration of the sample and control, respectively. K is the correction factor for the titer (oxalic acid), while D is the conversion factor for the tannin (for hydrolyzed—0.004157, condensed tannins—0.00582). Finally, V is the total volume (mL) of the extract, m represents the mass of the raw material, and V_3 is the volume (10 mL) of the extract obtained for the titration.

The content of the extractives was determined using gravimetric method, as described below. For the test pipette, we selected 25 mL of the final test extract; transferred it to one that was previously dried at a temperature of 100–105 °C to constant weight; accurately weighed a porcelain cup with a diameter of 7–9 cm; and evaporated it in a water bath until it reached dryness. The cup with the residue was dried at a temperature of 100–105 °C to constant weight, then cooled for 30 min in a desiccator, at the bottom of which was anhydrous calcium chloride, and weighed. The content of extractives in percent (X_E) in respect to the dry material can be expressed as:

$$X_E = (m \cdot 100)/(m_1 \cdot (100 - W)), \quad (3)$$

where m and m_1 are the masses (g) of dry residue after drying, and the sample extract before drying, respectively. W represents the mass loss once the sample has completely dried.

The antioxidant activity of the extracts and samples of the cosmetic emulsion with the addition of extracts was determined using the Oyaizu method (FRAP method) [28]. A total of 0.2 mL of the sample was mixed with phosphate buffer (0.5 mL; 0.2 M; and pH 6.6) and potassium ferricyanide [$K_3Fe(CN)_6$] (0.5 mL; 1% solution). The resulting mixture was kept at 50 °C for 20 min, then 0.5 mL of a 30% solution of trichloroacetic acid was added to the mixture and filtered. To 0.5 mL of the obtained filtrate was added ferric (III) chloride $FeCl_3$ (0.1 mL, 0.1% solution). The optical density was determined using a ULAB 102 spectrophotometer (Shanghai Metash Instruments Co., Ltd., Shanghai, China) at a wavelength of 700 nm. The increase in the absorption of the reaction mixture indicates an increase in the ability of the samples to reduce ferric iron ($Fe^{3+} \rightarrow Fe^{2+}$). A 20% solution of ascorbic acid was used as a reference solution.

3. Results and Discussion

Preliminarily investigations on the physicochemical properties of the selected polyorganosiloxanes were carried out. The results were compared with the traditional extractants (water and 70% of aqueous alcoholic solution), as shown in Table 2.

Table 2. Physico-chemical parameters of traditional extractants and investigated polyorganosiloxanes.

Indicator, Unit of Measurement	Traditional Extractants		Polyorganosiloxanes					
	Distilled Water	70% Aqueous Alcoholic Solution	BRB CM 50	BRB PTM 20	Silicone Oil 350 cSt	BRB 526	BRB 1834	BRB 1288
Appearance	colorless, clear liquid		colorless, clear liquid		colorless, opaque, viscous liquid			white liquid
Refractive index at 25 °C	1.333	1.363	1.397	1.460	1.403	1.460	1.403	1.481
Density at 25 °C, g/cm^3	1.000	0.932	0.958	0.988	0.972	1.072	0.960	0.992
Viscosity at 25 °C, mm^2/s	0.89	2.52	4.00	22.50	367.50	260.00	6000.00	5.00
Surface tension at 22 °C, $\times 10^{-3}$ N/m	72.40	30.96	18.54	21.89	21.10	18.81	19.10	27.12
Cosine of the edge angle of wetting, cos θ	0.121 (83°)	0.648 (50°)	0.971 (14°)	0.891 (27°)	0.945(19°)	0.941 (19°)	0.280 (73°)	0.800 (36°)
Relative spreading, mm^2/10 min	70.85	86.55	660.19	580.77	482.81	103.82	195.97	298.50

Table 2 shows that the studied polyorganosiloxanes possess low surface tension (from 18.54 to 27.12·10^3 N/m) compared with those of a water–alcohol mixture (30.96·10^3 N/m)

and water ($72.40 \cdot 10^3$ N/m). Compared to traditional extractants, silicones are characterized by high degrees of spreading and wetting. These characteristics are crucial for the extraction process.

Then, we determined the qualitative (Table 3) and quantitative (Table 4) composition of silicone extracts of calendula and wormwood. We focused on the detection and quantification of biologically active molecules, including vitamins, flavonoids, tannins, terpenoids, and glycosides (Tables 3 and 4).

Table 3. Qualitative determination of biologically active substances in silicone extracts of calendula (c) and wormwood (w). (+ presence, − absence).

Substance	Polyorganosiloxanes											
	Silicone Oil 350 cSt		BRB CM 50		BRB PTM 20		BRB 526		BRB 1834		BRB 1288	
	c	w	c	w	c	w	c	w	c	w	c	w
B_1, thiamine	+	+	+	+	+	+	+	+	+	+	+	+
B_2, riboflavin	−	−	−	−	−	−	−	−	−	−	−	−
B_5, pantothenic acid	+	+	+	+	+	+	+	+	+	+	+	+
B_6, pyridoxine	−	−	−	−	−	−	−	−	−	−	−	−
PP, nicotinamide	+	+	+	+	+	+	+	+	+	+	+	+
P, routine	+	+	+	+	+	+	+	+	+	+	+	+
C, ascorbic acid	+	+	+	+	+	+	+	+	+	+	+	+
A, retinol	−	−	−	−	−	−	−	−	−	−	+	+
E, tocopherol	−	−	−	−	−	−	−	−	−	−	+	+
General reactions to:												
–flavonoids	+	+	+	+	+	+	+	+	+	+	+	
–tannins	+	+	+	+	+	+	+	+	+	+	+	+
–glycosides	−	−	+	+	−	+	+	+	+	+	+	+

According to Table 3, it can be argued that the studied polyorganosiloxanes are able to extract biologically active substances from plant raw materials of calendula and wormwood. The presence of vitamins A and E in the samples of silicone extract taken from amodimethicone BRB 1288 should be noted. These results can be explained by the presence of cetrimonium chloride and tridecet-12 in the mixed polymer BRB 1288 surfactants, which significantly increase the wetting and permeability of silicone. The absence of glycosides in the silicone extracts of wormwood Silicone Oil 350 cSt and BRB PTM 20 can be related to the specificity of the chemical structure of these silicones. Thus, it can be concluded that the studied polyorganosiloxanes exhibited excellent extraction properties for biologically active substances contained in plants.

Table 4. Quantitative content of biologically active substances in silicone extracts of calendula (c) and wormwood (w).

Substance	Aqueous Extract		Polyorganosiloxane											
			Silicone Oil 350 cSt		BRB CM 50		BRB PTM 20		BRB 526		BRB 1834		BRB 1288	
	c	w	c	w	c	w	c	w	c	w	c	w	c	w
Vitamin C, g/L	13.20	8.80	0.60	0.52	2.00	3.20	2.71	0.50	113.70	164.90	3.62	2.11	15.72	41.91
The quantity of: flavonoids, %	5.09	0.61	13.71	16.29	13.14	13.89	24.51	24.89	23.87	19.85	13.93	13.46	45.12	40.48
Tannins, %	0.73	1.45	0.52	0.34	0.31	0.15	0.26	0.24	1.69	1.39	0.68	0.25	0.59	0.65
Extractives, %	0.10	0.10	2.09	3.12	2.11	3.10	2.21	3.32	2.52	3.20	2.26	3.43	3.03	3.98

Table 4 shows that the use of one-component silicones (polydimethylsiloxane Silicone Oil 350 cSt, cyclopentasiloxane BRB CM 50, and phenyltrimethicone BRB PTM 20) and a mixed solution of dimethiconol in cyclopentasiloxane BRB 1834 does not promote the extraction of vitamin C from raw materials. The use of amodimethicone silicone BRB 1288 allows for the extraction of vitamin C (15.72 g/L) with an efficiency similar to the aqueous

extract (13.20 g/L). The extraction using PEG-12 polydimethylsiloxane BRB 526 drives to significantly increase the content (113.70 g/L) of vitamin C in the extract relative to the aqueous extract (13.20 g/L).

The effects of polyorganosiloxanes on the extraction of vitamin C from wormwood raw materials are similar to those of calendula.

All the studied polyorganosiloxanes, unlike water, were able to extract significant amounts of flavonoids from the calendula and wormwood (Table 4). This represents an advantage compared to the process of extraction using aqueous solvents. The quantity of flavonoids of silicone extracts in comparison with those of water were increased by 2.6–8.8 and 22.1–66.4 times for calendula and wormwood, respectively. The best results in the content of flavonoids (45.12% for calendula and 40.48% for wormwood) were obtained by using amodimethicone BRB 1288, which contains aminoethylaminopropylsiloxane, cetrimonium chloride, and tridecet-12.

Tannins are able to seal cell membranes, creating a barrier from harmful factors. They are synergists for substances that exhibit antioxidant properties. Such functions of polyphenols are preserved in the composition of cosmetic products, when applying cosmetics to the skin [29].

The obtained research results (Table 4) indicate that the content of tannins in the silicone extract of calendula using PEG-12 polydimethylsiloxane BRB 526 is 1.685%, which is 2.3 times higher than the quantity for aqueous extract (0.730%). The use of a solution consisting of dimethiconol in cyclopentasiloxane BRB 1834; amodimethicone BRB 1288; and polydimethylsiloxane Silicone Oil 350 cSt, allows for the attainment of 0.681, 0.594, and 0.519% of tannins, respectively. The lowest values were observed for cyclopentasiloxane BRB CM 50 (0.314%) and phenyltrimethicone BRB PTM 20 (0.257%). Similar results were estimated for the silicone extracts obtained from wormwood raw materials.

As shown in Table 4, the highest contents of extractives for both calendula (3.034%) and wormwood (3.988%) were obtained by using the extractant amodimethicone BRB 1288. The other polyorganosiloxanes exhibited fairly high rates of extraction. More significantly, the extractive contents ranged from 2092% to 2516% for calendula and from 3101% to 3430% for wormwood. It is interesting to note that aqueous extracts of calendula and wormwood contained only 0.100% of extractives. The results of a comprehensive study of the qualitative and quantitative contents of the main biologically active substances suggest that the selected polyorganosiloxanes can be used as extractants, and the obtained extracts should be offered as ingredients of complex action for emulsions for cosmetic purposes. Since biologically active substances possess specific functions and properties, it is interesting to investigate their antioxidant action, which is crucial for several functions in biotechnological applications.

The antioxidant activity of samples of silicone plant extracts was determined by the Oyaizu method (FRAP method). The obtained results are presented in Table 5.

Table 5. Antioxidant activity of the silicone extracts.

Extract	Solution of Comparison	Aqueous Extract	Polyorganosiloxanes					
			Silicone Oil 350 cSt	BRB CM 50	BRB PTM 20	BRB 526	BRB 1834	BRB 1288
	Optical Density, D							
Calendula	0.770	0.711	0.457	0.221	0.218	>3	1.507	>3
Wormwood		0.657	0.451	0.254	0.204	>3	1.111	>3

After analyzing the obtained data (Table 5), it can be noted that the plant extracts based on mixed quantities of polyorganosiloxanes PEG-12, polydimethylsiloxane BRB 526, and amodimethicone BRB 1288 are characterized by the highest antioxidant activity (>3 compared to 20% of the ascorbic acid solution, which is 0.770). The use of a solution of dimethiconol in cyclopentasiloxane BRB 1834 also allows for the attainment of high rates

for the calendula and wormwood plant extracts (1.507 and 1.111, respectively). The use of one-component silicones of polydimethylsiloxane Silicone Oil 350 cSt, cyclopentasiloxane BRB CM 50, and phenyltrimethicone BRB PTM 20 does not permit the achievement of high levels of antioxidant activity. The values range between 0.204 and 0.457, which is lower than those obtained for the aqueous extracts of calendula (0.711), wormwood (0.657), and the reference solution (0.770).

Based on these results, we can conclude that the extracts obtained using one-component silicones are characterized by a low antioxidant activity, although they contain biologically active substances. The mixtures of polyorganosiloxanes exhibited a better extraction capacity enhancing the antioxidant activity of the extracts. Accordingly, the obtained silicone plant extracts of biologically active substances can be used in the emulsions for cosmetic purposes. The relevant efficiency of extraction using polyorganosiloxanes can be explained by their physicochemical properties, including surface tension, fluidity, and the partition coefficient. Due to these characteristics, they penetrate better and faster through cell membranes leading to the extraction of biologically active substances. The polyorganosiloxanes mixtures evidenced a greater extraction efficiency because of the presence in their composition of two surfactants (BRB 526 and BRB 1288) and the polar -OH group in polydimethylsiloxanol (BRB 1834). These components contributed to the enhancement of the extraction process and provided a wider complex of biologically active substances.

4. Conclusions

We investigated the efficiency of several polyorganosiloxanes (single components and mixtures) as extractants of biologically active substances contained in vegetable raw materials (*Calendula Officialis* L. and *Artemisia Absinthium* L.). Compared with the traditional extractants (water and water/alcohol mixtures), the investigated polyorganosiloxanes can be more efficient in the extraction of biological molecules, as suggested by their physicochemical properties. In addition, polyorganosiloxanes do not require the removal step after the extraction process. The latter represent a greater advantage with respect to the use of the traditional extractants.

The results of a comprehensive study on the qualitative and quantitative contents of biologically active substances highlighted that the selected polyorganosiloxanes are promising as extractants, and the resulting extracts can be employed as ingredients of complex action for emulsions used for cosmetic purposes.

Based on the determination of the antioxidant activity of the obtained silicone plant extracts, we observed that mixtures of polyorganosiloxanes amodimeticone BRB 1288, PEG-12 polydimethylsiloxane BRB 526, and a solution of dimethiconol in cyclopentasiloxane BRB 1834 were the most effective extractants. This paper shows that the combination of different polyorganosiloxanes reveals a route for the development of novel protocols in the extraction of biologically active molecules from plants.

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