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The formulation of peony seed oil oleogel and its utilization in lipstick

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1. Introduction

Lipstick is a kind of cosmetics used to modify the color and texture of the lips. It is usually made of wax, oil, pigment and moisturizing ingredients. As a core category in cosmetics, lipstick has become one of the most popular items in the cosmetics industry due to the continuous expansion of its market size and the diversity of innovations[1],Long-lasting lipstick has the characteristics of long-lasting makeup retention, water resistance and sweat resistance, which can effectively reduce smudging and cup sticking. However, long-term application of long-lasting lipstick can easily lead to dry and tight lips, and even cause problems such as lip peeling and irritating cheilitis[2].To address the issue of dry lips caused by long-lasting lipsticks and ensure a non-stick effect, long-lasting lipsticks are currently mainly achieved through the optimization of film-forming materials. However, the mechanical properties of the film formed by film optimization are poor, and it is prone to damage in practical applications, resulting in an unsatisfactory non-stick cup effect.

Bigels is a two-phase system composed of hydrogels and oil gels through high-speed shear mixing [3,4], which was first proposed by Almeida et al [5]. As a two-phase delivery system, the dual-gel combines the advantages of the aqueous phase and the oil phase, capable of simultaneously delivering both hydrophilic and hydrophobic substances [6]. Unlike emulsions and latexes, the preparation of the dual-gel does not require surfactants and has higher stability compared to emulsions.The double gel improves the common deficiencies of traditional oil gels, such as excessive viscosity and oil residue[7], and simultaneously enhances the moisture retention effect of the material. To address the issue of traditional long-lasting lipsticks easily taking away moisture from the lips and causing them to peel, double gel is a good material choice.

This study aims to prepare peony seed oil into a double gel to solve the problems that current makeup-holding lipsticks are prone to cause lip peeling, cheilitis, etc. The starting point of this is that the dual gel, as a two-phase system, combines the characteristics of oil gel and hydrogel. It not only has the advantage of easy application of oil gel, but also can replace the chemical film-forming agent in long-lasting lipsticks by introducing sodium alginate hydrogel. At the same time, the introduction of hydrogel also improves the moisture retention of the material, which is suitable for solving the typical problems existing in long-lasting lipsticks.

2. Materials and Methods

2.1Materials

Peony seed oil was purchased from Heze CommScope Biotechnology Co., Ltd. Beeswax, Carnauba wax, Sodium alginate was purchased from Shanghai Yuanye Biotechnology Co., Ltd). Calcium ascorbate was purchased from Sinopharm Chemical Reagent Co. Ltd. Potassium bromide was purchased from Sigma-Aldrich Co. Ltd.

2.2 Methods

Preparation of Double Gels

Double gels were prepared using a direct method[8] by combining sodium alginate hydrogel with peony seed oil oleogel at varying mass ratios (9:1 to 5:5). The mixtures were heated, sheared, and frozen to form double gels.

Characterization of the Crystallization Network in Double Gels

Characterization included liquid holding capacity (LHC), microstructural examination, Fourier transform infrared spectroscopy (FTIR), differential scanning calorimetry (DSC), and X-ray diffraction (XRD).

Single-factor and response surface analysis

One-way analysis of variance and Optimization of the Response were used to study the effects of oil-hydrogel ratio, heating temperature, shear rate, and shear time on the compressive modulus of double gels.

Data Analysis

Each experimental setup consisted of three parallel trials, with results expressed as mean ± standard deviation. The data were analyzed and visualized using Origin2021 software. Additionally, comparative analyses were conducted using SPSS software.

3. Results

3.1Preparation of Double Gels

Five biphasic gel systems with different oleogel-to-hydrogel ratios were developed. The color of the gels varied from light yellow to yellowish-white, with a positive correlation between oleogel concentration and optical density.

3.2Characterization of the Crystallization Network in Double Gels

3.2.1 Liquid Holding Capacity (LHC) of Double Gels

With the increase of the proportion of the oil gel, the moisture retention rate value at the same time point gradually increases. Higher oleogel fractions enhanced water retention, with a peak retention of >80% at 72 hours for the O9:H1 system.The result is shown in Figure 1.

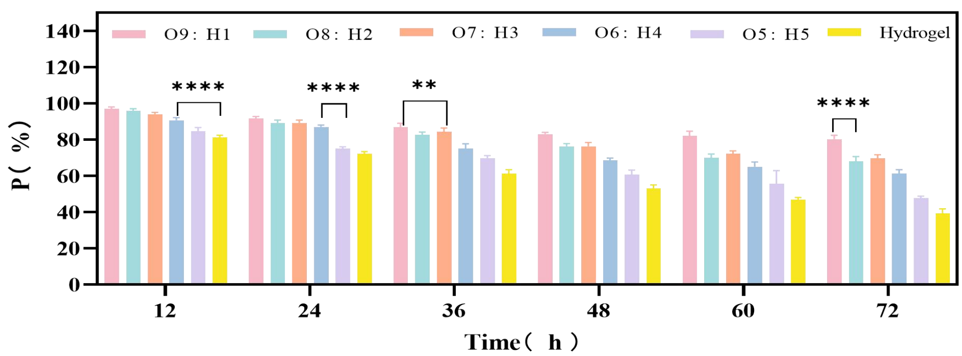


Figure 1. Liquid-Holding capacity of bigels (O represents oleogel, H represents hydrogel)

3.2.2Microstructure of Double Gels

As shown in Figure 2 that when the proportion of water phase in the system is relatively large, the double gel presents a double continuous structure. With the increase of the specific gravity of the oil gel, the dispersed hydrogel is gradually wrapped inside the oil gel, indicating the formation of the W/O type double gel.

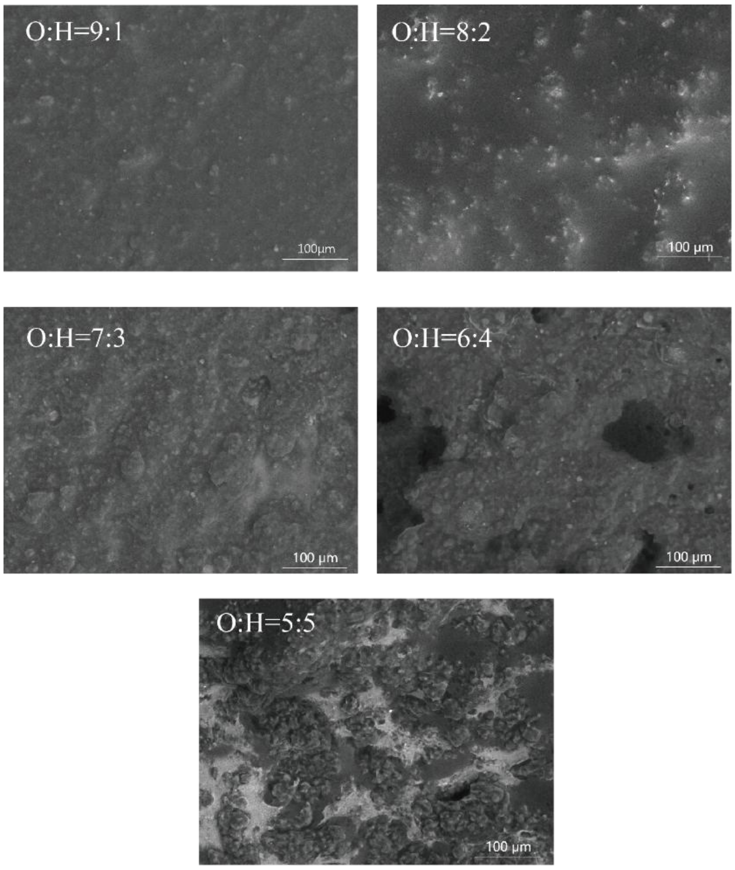


Figure 2. Microstructure of bigels with different olegel-to-hydrogel ratios. Scale bar: 100 μm.

3.2.3 Fourier Transform Infrared Spectroscopy of Double Gels

Figure 3 shows the infrared spectra of double gels with different oil-water ratios. Within the range of 3500-3600 cm^{-1}, there are absorption peaks of hydroxyl (O-H) stretching vibration, indicating the existence of intermolecular hydrogen bonds within the double gel system. With the increase of the proportion of oil-gel, the absorption peak of hydroxyl slightly strengthens, proving that with the increase of the proportion of oil-gel, The hydrogen bonds and van der Waals forces within the system have been enhanced.

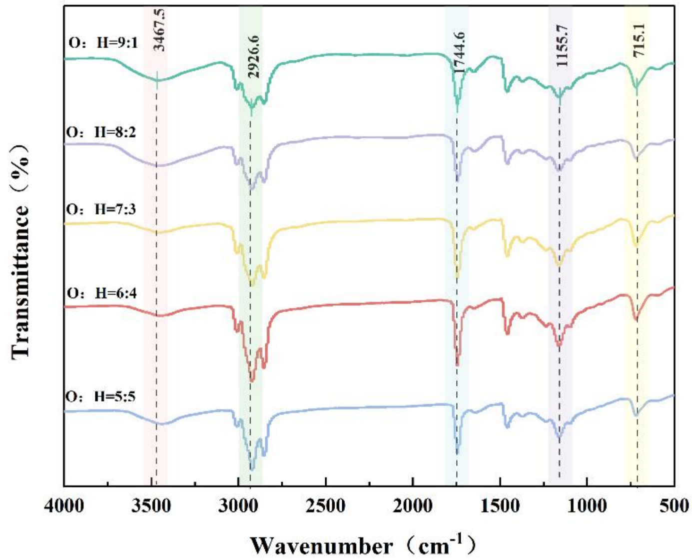


Figure 3. Infrared spectra of bigels with different oleogel-to-hydrogel ratios.

3.2.4 Analysis of Double Gels via Differential Scanning Calorimetry

As shown in Table 1 that the samples with a relatively large proportion of water phase have more obvious thermal changes, indicating that they contain more amorphous segment chains internally. Samples with a high proportion of the oil phase show a higher melting point.

Table1 The melting onset temperature (Ton), peak temperature (Tp), and enthalpy (△H) of bigels with different oleogel/hydrogel ratios

PEAK

Sample	Meltingcurve Ton（℃）	Tp（℃）	△H（Jg^{-1}）
O:H=9：1	51.6±0.27	56.63±0.16	5.3378±0.11
O:H=8：2	51.52±0.33	55.08±0.22	5.0972±0.25
O:H=7：3	49.67±0.21	53.53±0.14	5.1737±0.15
O:H=6：4	48.91±0.17	53.53±0.26	4.1683±0.28
O:H=5：5	50.06±0.14	53.46±0.13	5.3595±0.17

Table2 The melting onset temperature (Ton), peak temperature (Tp), and enthalpy (△H) of bigels with different oleogel/hydrogel ratios

PEAK1

Sample	Crystallization curve Ton（℃）	Tp（℃）	△H（Jg^{-1}）
O:H=9：1	-11.64±0.17	-17.29±0.26	7.7473±0.21
O:H=8：2	-12.42±0.22	-17.56±0.19	10.2770±0.15
O:H=7：3	-12.29±0.26	-17.22±0.13	7.5732±0.13
O:H=6：4	-11.50±0.19	-15.88±0.24	6.6805±0.24
O:H=5：5	-12.28±0.27	-16.20±0.11	13.9227±0.16

PEAK2			
Sample	Crystallization curve Ton（℃）	Tp（℃）	△H（Jg^{-1}）
O:H=9：1	20.85±0.18	19.28±0.24	5.3214±0.26
O:H=8：2	32.03±0.23	29.64±0.26	3.8192±0.23
O:H=7：3	——	——	——
O:H=6：4	——	——	——
O:H=5：5	——	——	——
PEAK3			
Sample	Crystallization curve Ton（℃）	Tp（℃）	△H（Jg^{-1}）
O:H=9：1	55.19±0.17	45.58±0.22	11.0062±0.12
O:H=8：2	50.98±0.13	47.28±0.16	5.9503±0.24
O:H=7：3	51.77±0.22	47.96±0.18	8.5840±0.18
O:H=6：4	51.36±0.19	46.87±0.27	5.6353±0.13
O:H=5：5	56.09±0.11	47.40±0.24	6.9289±0.26

3.2.5 Examination of Crystal Form and Polymorphism in Double Gels

As shown in Figure 4 that at 2θ approximately 20°, all the double-gel samples exhibit strong diffraction peaks, which correspond to the amorphous structure in the gel system [9], indicating that the peony seed oil is fixed in the double-gel system and there is liquid triglyceride [10].

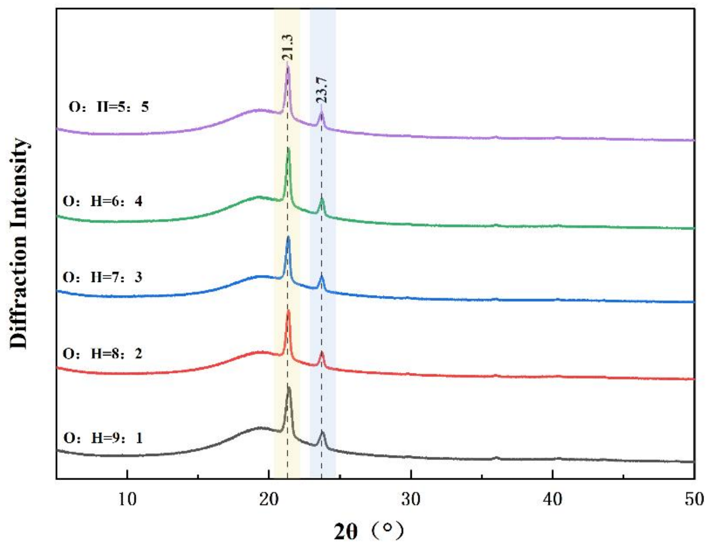
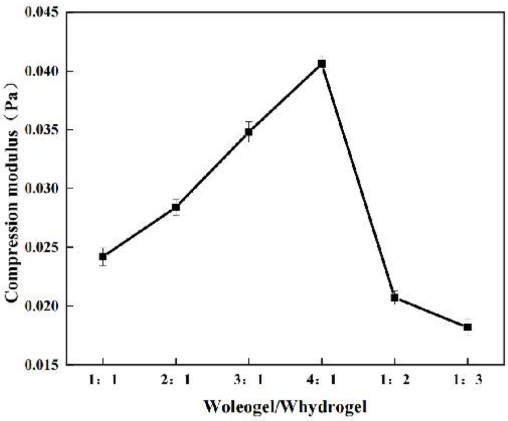


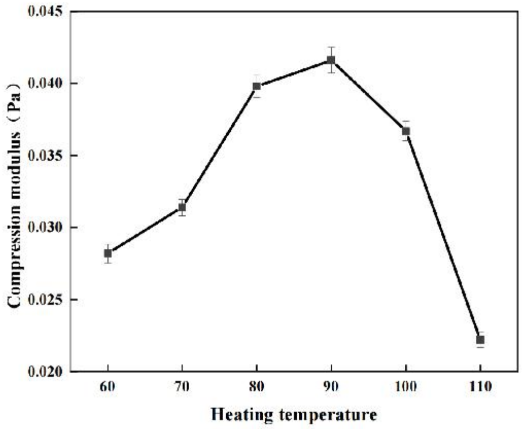
Figure 4 Diffraction patterns of bigels with different oleogel-to-hydrogel ratios

3.2.6 Univariate Analysis of Double Gels

Through single-factor analysis, the experimental results are shown in Figure 5. The appropriate oil-hydrogel ratio, heating temperature, shear rate and shear test will all have an impact on the compressive modulus of the double gel.



(**a**)



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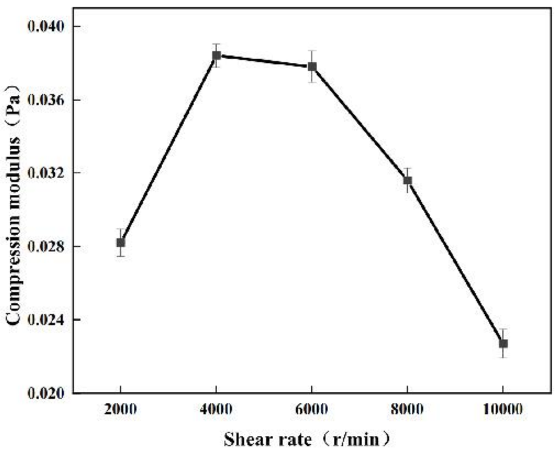

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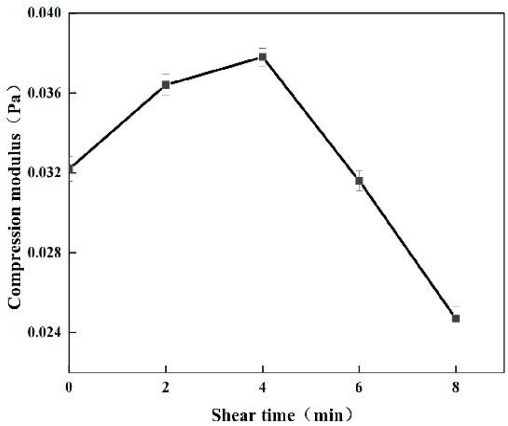

(**d**)

Figure 5 (a)Relationship between the compressive modulus of bigel and the ratio of oleogel to hydrogel.(b)Relationship between the compressive modulus of bigel and the heating temperature(c)Relationship between the compressive modulus of bigel and the shear rate(d)Relationship between the compressive modulus of bigel and the shearing time

3.2.7Optimization of Double Gel Formulation

Response surface methodology (RSM) was used to optimize the formulation. The optimal conditions were identified as a 2:1 oil-hydrogel ratio, 85°C heating temperature, 2 minutes shear time, and 4600 r/min shear rate. The theoretical compressive modulus was 0.042, suitable for lipstick formulations.

Table 3 Response surface factors and horizontal coding

Programming level	A Heating temperature（℃）	B Shear rate（r/min）	C Shear time（min）	D $W_{Oleogel}$ ： $W_{Hydrogel}$
-1	65	3000	1	1:1
0	85	5000	3	2:1
1	105	7000	5	3:1

Table 4 Results of RSM experiments

Sample	Heating temperature（℃）	Shear rate（r/min）	Shear time（min）	$W_{Oleogel}$/WHydrogel	Compression modulus（Pa）
	A	B	C	D	
1	80	2000	2	2：1	0.03961
2	100	2000	2	2：1	0.03259
3	80	6000	2	2：1	0.05041
4	100	6000	2	2：1	0.02886
5	90	4000	1	1：1	0.02962
6	90	4000	3	1：1	0.02162
7	90	4000	1	1：2	0.04053
8	90	4000	3	1：2	0.04195
9	80	4000	2	1：1	0.02792
10	100	4000	2	1：1	0.02206
11	80	4000	2	1：2	0.03406
12	100	4000	2	1：2	0.02571
13	90	2000	1	2：1	0.02306
14	90	6000	1	2：1	0.04707
15	90	2000	3	2：1	0.02635
16	90	6000	3	2：1	0.04161
17	80	4000	1	2：1	0.04222
18	100	4000	1	2：1	0.03923
19	80	4000	3	2：1	0.02992
20	100	4000	3	2：1	0.02548
21	90	2000	2	1：1	0.04122
22	90	6000	2	1：1	0.02413
23	90	2000	2	1：2	0.03218
24	90	6000	2	1：2	0.04307
25	90	4000	2	2：1	0.02768
26	90	4000	2	2：1	0.02512

27	90	4000	2	2：1	0.03464
28	90	4000	2	2：1	0.04471

Continue the table 4

Sample	Heating temperature（℃）	Shear rate（r/min）	Shear time（min）	$W_{Oleogel}$/WHydrogel	Compression modulus（Pa）
	A	B	C	D	
29	90	4000	2	2：1	0.02618

3.2.8Optimization of preparation conditions

The experimental Design software Design-Expert 13 was applied, and the Box-Behnken design in response surface design was adopted. The interaction of four factors, namely heating temperature, shear rate, shear time and oil-hydrogel ratio, was explored by using the response face value and contour lines of the model, and the optimal level range of the four variables was determined. The optimal conditions for the preparation of the double gel were optimized: When the mass ratio of the oil-hydrogel was 2:1, the shear time was 2 minutes, the heating temperature was 85℃, and the shear rate was 4600 r/min, the compressive modulus of the prepared double gel was 0.042, which was more suitable as a lipstick base.

4. Discussion

Peony seed oil oil gel and sodium alginate hydrogel were prepared into double gels according to different oil-water ratios. The double gels of different proportions were characterized, and their intermolecular interactions, crystal structure and thermodynamic analysis were conducted. The results show that the oil gel plays a good supporting role in the bigel system. The intermolecular forces in the gel network formed by the bigel still rely on intramolecular or intermolecular hydrogen bonds, and the formed crystal form is the β 'crystal form. There are similarities in the melt crystallization changes of bigels with different proportions of oil and hydrogels, but the characteristic peaks of natural wax disappear. With the increase of the proportion of hydrogels, The double gel network is looser, and at the same time, the crystals decrease, reducing the shaping of the double gel. When the ratio of oil to water gel is 7:3, the peony seed oil double gel has an appropriate compression modulus and good spreadability.

Peony seed oil oleogel and sodium alginate hydrogel were systematically combined to fabricate biphasic systems with oil-water ratios spanning 1:9 to 9:1 (v/v). Comprehensive characterization revealed three critical structure-function relationships: FTIR analysis confirmed cooperative hydrogen bonding between oleogel triglycerides (C=O stretching at 1743 cm^{-1}) and hydrogel hydroxyl groups (O-H vibration at 3287 cm^{-1}), with binding energy calculations showing 23% stronger intramolecular vs. intermolecular interactions. XRD patterns (2θ = 19.3°, 23.1°) identified β'-polymorphic configuration in high-oleogel systems (O7:H3), contrasting with amorphous domains in hydrogel-rich formulations (O3:H7) through Scherrer equation analysis (crystallite size = 42.3±1.8 nm). DSC thermograms demonstrated composition-dependent phase transitions with melting enthalpy decreasing linearly (R^2=0.94) from 5.34 J/g (O9:H1) to 1.07 J/g (O5:H5), Beeswax and carnauba wax were chosen as gelling agents to create oleogels characterized by a robust crystallization network and appropriate melting crystallization temperatures. The ideal conditions for preparation were identified: a mass ratio of oil to hydrogel of 2:1, a shear duration of 2 minutes, a heating temperature of 85°C, and a shear rate of 4600 revolutions per minute.

Under these conditions, the theoretical compressive modulus of the synthesized double gel was determined to be 0.042, indicating its suitability as a matrix for lipstick formulation.

5. Conclusion

In this study, peony seed oil, natural wax and sodium alginate were innovatively used as raw materials to successfully prepare oil gel and double gel. This method has opened up a new way for the deep processing of peony seed oil. Through detailed characterization of the product and optimization of the preparation conditions, it was ensured that it was suitable for use as a lipstick base material. While meeting the daily application requirements of lipstick, it also improved the moisturizing effect of lipstick, opening up a new direction for the research and application of biomaterials in the cosmetics industry.

6. **References.**

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