

New emulsion type liquid lip formulation with gloss and stain effect using shear stress-induced phase separation phenomenon

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Abstract

Recently, “emulsion type liquid lip” is getting attention in the color cosmetics market for lips. Emulsion type liquid lip is a type of lip color cosmetic formulation that has recently gained popularity. This formulation can create unique colors and textures.

However, there has been a limitation in that gloss and stain, one of the important properties of lip color cosmetics, has a trade-off relationship with each other in this formulation. The goal of this study is to develop an emulsion type liquid lip formulation that simultaneously enhances the stain effect and the gloss effect by using the phase separation phenomenon of the formulation. The phase separation of the formulation is caused by the shear stress. Shear stress with the appropriate strength can be easily created by users when they rub their lips. Therefore, users can easily create the phase separation phenomenon.

This formulation does not contain relatively high viscosity and high refractive index oil for gloss effect, unlike conventional formulations. Low viscosity, low density oils were applied to this formulation instead of it. In addition, we adopted unusual methods of controlling the pH and salinity of the formulation to induce a strong stain effect. As a result of the experiment, it was confirmed that the developed formulation had a superior stain effect than the conventional formulation and similar glossiness with the conventional formulation.

Several techniques were used to analyze the cause of the phase behavior of the formulation and to evaluate its effect. Rheological measurements were performed using a rotational rheometer. In addition, the physical and chemical properties of the experimental formulations were analyzed by moisture quantification, confocal laser fluorescence microscopy, gloss measurement, and others. Based on the results of this study in the future, it is expected that emulsion type liquid lip formulation can be developed more systematically.

Keywords: acid dye, DWS, phase separation, color cosmetics, glossiness

Introduction.

Recently, emulsion-type liquid lip formulations are attracting attention in the color cosmetics market. emulsion type liquid lip formulations make unique colors and sensory that are difficult to carry out in conventional formulations, such as lipsticks or liquid lips composed only with oil phases. However, cosmetical formulators find it difficult to control the gloss and color staining effects of emulsion-type liquid lip formulations as they wish. There is a limitation in that gloss and staining, which are the main makeup effects of color cosmetics for lips, are in a trade-off relationship with each other for emulsion-type liquid lip formulations. It is physically natural that the gloss effect and the staining effect of the emulsion-type liquid lip formulation are inversely proportional to each other.

The staining effect is mostly caused by adsorption of acid dye (also called azo dye), a water-soluble dye used in color cosmetics, to the stratum corneum.[1] Acid dye is stained with stratum corneum for several reasons.

The first staining mechanism is the electrical attraction binding of dyes dissociated into anions. Tregear, R. T., & Dirnhuber, P. did not directly apply acid dye to the *in vivo* skin in their study. [1] Stratum corneum was stripped with adhesive tape and keratin remaining on the tape was stained with dye, Carmoisine W. This dye has sulfonate groups ($-\text{SO}_3^-$) and is classified as acid dyes. The study claimed that keratin stained by acid dyes is caused by adsorption or loose chemical bonding. In particular, the cationic group in the acid dye bond with the protein of skin by the electrical attraction and plays a major role in adsorption.

The isoelectric point of keratin, the substance constituting the stratum corneum, is about pH 5. Therefore, the binding amount of acid dye molecule, which has a negative charge in a state of being dissolved in water, varies according to the pH of the stratum corneum.[2]

The second staining mechanism relates to the presence of salt such as sodium chloride. Acid dye is also called direct dye in the textile dyeing industry. Direct dye does not require a fixing process such as mordanting to dye fibers. However, sodium chloride can strengthen coloring like a mordant that enhances the staining effect. If sodium chloride is dissociated together as an electrolyte in an aqueous dye solution, it helps to stain. The reason why staining is strengthened is that sodium chloride has an effect that helps the 'salting-out' effect and adsorption to fibers.[3] This study case is for cellulose fibers and acid dyes, but it is thought that they are not different from the mechanism of acid dyes that are stained on the skin. The fact that both the proposed first and second staining mechanisms can occur when the acid dye is dissociated in water is a very important fact for this study.

The goal of this study is to enhance the staining effect and gloss effect based on the phase separation between the oil phase and the water phase. Because the water phase is located close to the skin, and the dye dissociated in the aqueous solution in this state has a larger contact area with the skin than the emulsion, it was expected that the adsorption between the stratum corneum and the dye would occur more.

The argument that the gloss effect and the staining effect of the emulsion-type liquid lip formulation are inversely proportional to each other can be interpreted based on the understanding of the skin staining mechanism of acid dye and gloss phenomenon.

How does the gloss work? To understand the gloss effect of makeup products on lips, it is necessary to know what gloss is. First of all, it is necessary to recognize that gloss is a perceptual definition, not a physical definition. In this paper, perceptual gloss by cosmetic formulation will be named 'gloss effect'. Some researchers had argued that gloss consists of specular reflection, a physical phenomenon.[4] Based on this argument, a patent was applied for a device for measuring surface gloss.[5]

However, the consensus is that gloss is a perception that is felt as a result of multidimensional influences of various factors. Factors that affect the gloss are the observer, lighting conditions, brightness, contrast, specular reflectance, surface texture, highlights and their properties, and specular mirror images.[4] Nevertheless, the ratio of diffuse and specular reflection is an important physical quantity proportional to the gloss. [6] Specular reflection can be calculated theoretically by the Fresnel equation. (Equation 1) [7]

$$R_s = \frac{1}{2} \left[\left(\frac{\cos i - \sqrt{m^2 - \sin^2 i}}{\cos i + \sqrt{m^2 - \sin^2 i}} \right)^2 + \left(\frac{m^2 \cos i - \sqrt{m^2 - \sin^2 i}}{m^2 \cos i + \sqrt{m^2 - \sin^2 i}} \right)^2 \right] \quad \text{Equation 1}$$

$$R_r = R_s \cdot \exp \left(- \left(\frac{4m\sigma}{\lambda} \cdot \cos i \right)^2 \right) \quad \text{Equation 2}$$

Where R_s is specular reflectance, R_r is specular reflectance of a medium of refractive index m bounded by a rough surface [8], [9], m is the refractive index of the surface specimen, i is the angle of incidence, σ is roughness and λ is wavelength.

Based on the understanding of the Fresnel equation, the factors affecting the gloss effect of the formulation are as follows. The first factor is the refractive index of the oil. Oil with a high refractive index has a lot of specular reflection according to the Fresnel equation. Another factor is the smoothness of the surface. The smoother the surface formed when the formulation is applied to the lips, the lower the diffuse reflection of light and the higher the reflection.

Some studies have shown that the gloss effect of lipstick can be controlled by the properties of raw materials based on a physical understanding of reflection. [10] de Clermont-Gallerande *et al.* confirmed by experiment that the refractive index of the oil is proportional to the gloss value of the oil raw material measured by the instrument. However, the viscosity of the oil had little to do with the gloss measured by the instrument.

Surprisingly, according to the evaluation of the gloss effect by the sensorial analysis by panelists, the refractive index of oil materials does not affect the gloss effect felt by humans for each lipstick formulation. The hypothesis for the gloss effect of the lipstick presented in this study [10] is that smooth and uniformly coated lipstick with a constant thickness results in a larger specular reflection than when the lipstick is roughly coated. Therefore, the degree to which the surface of the applied lipstick is smooth enhances the gloss effect of the lipstick. Also, if a high-viscosity oil is included in the lipstick formulation, the thickness of the lipstick applied to the lips becomes thicker. Wax, one of the main raw materials that compose lipstick, is usually in a solid crystalline state. Wax has the property of reducing specular reflection by scattering light. On the other hand, since the oil exists in a liquid state, the higher the content of the oil, the smoother the surface of the lipstick-coated lips. Therefore, the high viscosity oil not only improves the mechanical performance of preventing the lipstick from breaking easily but also improves the gloss effect because the lipstick does not flow well from the surface of the lips and creates a smooth surface condition with a uniform thickness.

The lipstick formulation is a single-phase formulation without water, and due to the nature of the formulation that needs to maintain the solid form, a lot of wax is used. Therefore, the emulsion-type liquid lip formulation, which is the formulation to be covered in this study, and the mechanism of generating the gloss effect of the lipstick cannot be completely the same.

Formulators mainly use polybutene raw materials to give emulsion-type liquid lip formulations a glossy effect. Polybutene is an oil with high viscosity properties. As described in the gloss mechanism of the lipstick previously, the high viscosity oil helps to form a smooth surface to impart a gloss effect. However, this formulation may be sticky and has a limitation in that it is not stained well. It is presumed that the reason why the coloring is not good is that acid dye, a water-soluble dye, is trapped in an emulsion structure that is not easily destroyed due to its high viscosity in formulations containing high viscosity oil and is less likely to be adsorbed to the skin of the lips.

From this point of view, this research tries to induce a gloss effect by allowing the emulsion to be phase-separated quickly by shear stress so that acid dye was adsorbed a lot on the skin, and also, the oil layer rose upward by density difference to form a smooth surface. [11] Tomoko Ikeda used the phase separation phenomenon to coat an oil film on the lip surface to barely leave a color mark on cups. However, the study differs from this study in that it was a study on lipstick formulations and did not mainly deal with staining or gloss effects.

Materials and Methods.

Although the INCI names used in the experimental formulation and the control formulation, which are the formulation used as controls, have not been disclosed, the approximate compositions are listed in Table 2. The dyes used in the formulations are D&C Red No.33, D&C Red No.28, and FD&C Yellow No.6. (Table 1) The refractive index of the oil mixture used in the experimental formulation is 1.43 and the viscosity is 10.6 mPa·s.

Table 1 Three acid dyes used in the liquid lip formulations.

Name from FDA color additive list	D&C Red No.33	D&C Red No.28	FD&C Yellow No.6
CAS No.	3567-66-6	18472-87-2	2783-94-0
Color Index Number	CI 17200	CI 45410	CI 15985
Molecular structure			

Table 2 Composition table of experimental formulation and control formulation

Control formulation (common O/W emulsion)		Experimental formulation		Modified experimental formulation [†]
Name	wt%	Name	wt%	wt%
Water	43.4	Water	39.3	39.5
surfactants	4	Surfactants	2.5	2.5
Silicone oil (high viscosity)	40	Silicone oil (low viscosity)		
branched-chain fatty alcohol		Ester oil	38	38
Lipophilic thickening agent	2.5	branched-chain fatty alcohol		
Hydrophilic polyacrylate Thickening Agent	5	Hydrophilic polyacrylate Thickening Agent	2.5	2.5
preservative	2.4	Lipophilic thickening agent	0	2.5
Acid dyes	2.7	Polyol	12	12
		Preservative	3	3
		Acid dyes	2.7	0

[†] No dye was added as it was a formulation for DWS measurement.

A glossmeter (micro-TRI-gloss, BYK) and high-resolution photographs (VISIA-CR, Canfield) were used to measure the gloss effect of the formulations. In order to check the

gloss effect trend of the experimental formulation and the control formulation using a glossmeter, the two formulations were applied in equal amounts on light brown artificial leather. And the glossiness (arbitrary unit: Gloss Unit) before rubbing the applied formulation and the glossiness after rubbing was measured. (Figure 1) To analyze the gloss effect using photographs, the experimental formulation was applied to the left side of the lips of a woman in her 30s, and the control formulation was applied to the right side. After the two formulations were applied to the lips, they rubbed each other to create friction. And the lip area was photographed using VISIA-CR.

A UV/Vis/NIR spectrometer (UV/VIS/NIR spectrometer, Avantes) was used to measure the staining effect of the formulations. At this spectrometer, an integrating sphere equipped with a halogen lamp is connected to facilitate the measurement of the skin reflection spectrum. In order to confirm the tendency of the skin staining effect of the experimental formulation and the control formulation, the formulations were applied to the forearm of an adult male in his 30s. The formulations were dropped in a volume of 60 μ l and they were spread in a circle with a diameter of 5 cm. And after leaving the formulations for 2 minutes, the area where the formulation was applied was wiped off with cleansing tissue. Then, the reflection spectrum in the range of 400 ~ 800 nm was measured with a UV/VIS/NIR spectrometer.

As a result of the pre-experiment, it was confirmed that the mass decreased due to moisture evaporation when the experimental formulation was phase-separated by shear stress. The moisture reduction trend was confirmed using a microbalance and the Karl Fisher Electrometry method (V20 Compact Volume KF Titer, Mettler Toledo).

Rotational rheometer (MCR 92, Anton Paar) and diffusing wave spectroscopy (DWS) (DWS Rheolab II, LS Instruments) were used to rheologically analyze the phase separation phenomenon mechanism of the experimental formulation. The change in rheological properties during the phase transition process of the experimental formulation was measured with a rotational rheometer. The geometry used to measure was a sandblasted parallel plate with a diameter of 25 mm. The height of the gap between the parallel plate was set to 1 mm. The behaviors of emulsion in experimental formulations and control groups (control formulation and modified experimental formulation) were analyzed using DWS. To facilitate DWS measurement, the formulation used for this measurement was made without any dye.

Also, confocal laser fluorescence microscopy (CLFM) (FV3000, Olympus) was used for observing the phase separation phenomena by shear stress. In the experiment using CLFM, two fluorescent dyes were used instead of the acid dyes used in the experimental formulation. The fluorescent dyes used were Nile red (emission: 585 nm, excitation: 509 nm) and Nile blue (emission: 660 nm, excitation: 630 nm). Nile red was dissolved in the oil phase of the formulation, and Nile blue was dissolved in the water phase.

To identify the phenomenon of phase separation by shear stress with CLFM, a three-dimensional fluorescence image of the formulation was first taken before shear stress was applied. Then, the formulation placed on the slide glass was rubbed to induce phase separation. The phase-separated formulation was photographed again in the same manner.

In order to check whether the pH affects the skin stains of the three types of acid dyes, nine aqueous solutions of acid dyes with various pH adjustments were prepared at a concentration of 0.1 w/v%. To check whether the salinity of NaCl affects the staining effect of the acid dye molecules on the skin, 0.66 w/v% NaCl aqueous solution was prepared one by one for each acid dye type. In addition, three types of acid dyes were added to 1,3-butylene glycol to prepare an acid dye solution dissolved or dispersed in polyol. 15 solutions of acid dyes were

dripped onto the back of the hand of an adult male in his thirties to visually check the staining effect and its intensity.

Results.

The same amount of experimental formulation and control formulation was applied to artificial leather and the glossiness (Gloss Unit) was measured with a glossmeter. (Figure 1) The control formulation contains a lot of high-viscosity oil to induce a gloss effect. Therefore, upon application of this formulation, an immediate high gloss effect occurs. However, rubbing did not enhance the gloss effect of the control formulation. The experimental formulation had lower Gloss Units compared to the control formulation immediately after application. However, after rubbing, the measured Gloss Unit increased about 1.7 times compared to before rubbing. If we compare the gloss effect of the two formulations after rubbing, the gloss of the two formulations is induced to a very similar degree.

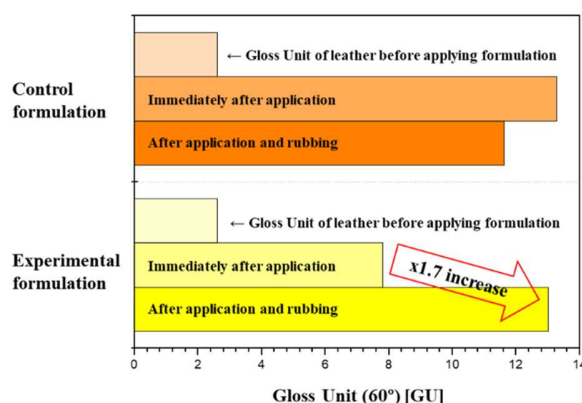


Figure 1 Glossiness results of the two formulations measured by glossmeter

This trend of gloss effect was also confirmed with VISIA-CR, an image analysis equipment. (Figure 2)

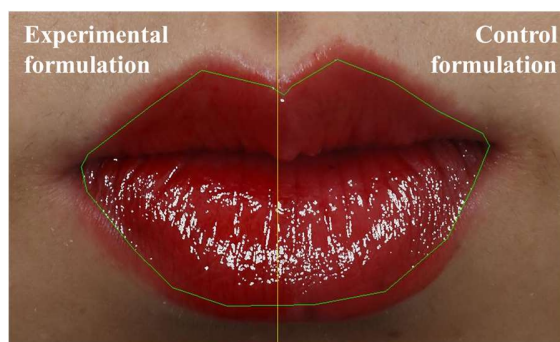


Figure 2 Gloss effect image analysis

The tendency of the staining effect of experimental formulation and control formulation was confirmed by UV/Vis/NIR spectrometer. In order to confirm the staining effect with the reflection spectrum, the reflection spectrum of the skin before coated was subtracted from the reflection spectrum of the same area washed just 2 minutes after coated. (Figure 3)

The reflectance in the wavelength over 600 nm increased in the washed skin after spreading compared to the bare skin before spreading of the experimental formulation. And the reflectance in the wavelength of less than 600 nm decreased. However, this tendency did not appear when the control formulation was applied. This result quantitatively proves that the skin could be stained red when the experimental formulation is applied to the skin.

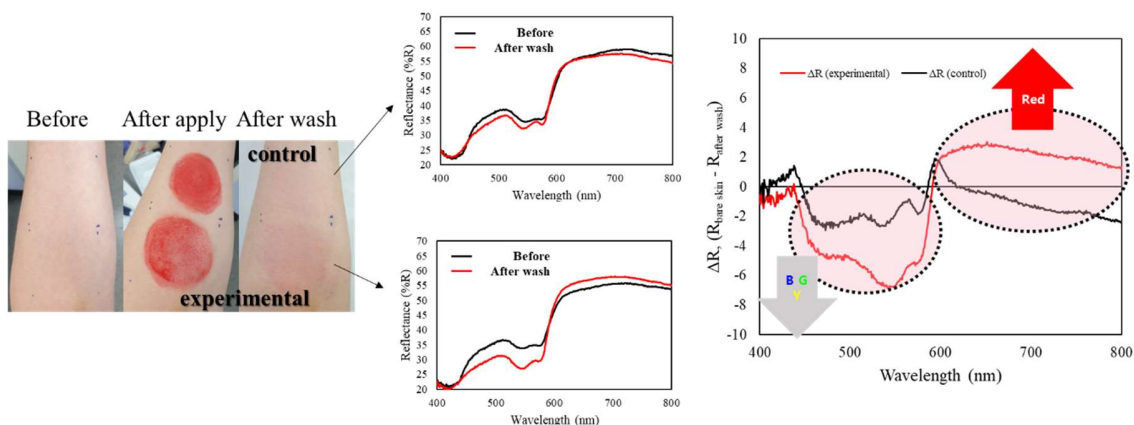


Figure 3 Reflection spectrum measurement before application and after washing

It was confirmed through the previous experimental results that the gloss effect was similar and the staining effect was excellent compared to the control formulation. Here, the results of analyzing the reason why the gloss effect and staining effect of the experimental formulation are excellent will be shown.

As a result of the preliminary experiment, when the experimental formulation is applied and rubbed with a finger, the oil layer is phase-separated into the upper layer. We tried to figure out why the experimental formulation was able to be separated and the control formulation was not possible. We tested whether there was a change in mass when this formulation was phase-separated. The experimental formulation was applied to the PMMA plate in an amount of 2 mg/cm². The PMMA plate was maintained at 32 °C by the heating stage. Its weight change was measured before and during the rubbing process. The same experiment was repeated three times, and in all experiments, it was confirmed that the phases were separated when the mass decreased by about 35 to 40%.

In order to determine what ingredient evaporated during the rubbing process, samples were collected while evaporating the experimental formulation in a reduced pressure environment. The reason for evaporating substances in a reduced pressure environment is to simulate an environment in which substances are evaporated without the influence of heat, similar to the real rubbing situation. The aliquoted sample was measured for water content by the Karl Fischer electrometric titration method. As a result, it was confirmed that most of the evaporated material was water. (Figure 4) The control formulation only reduced the mass by 26%.

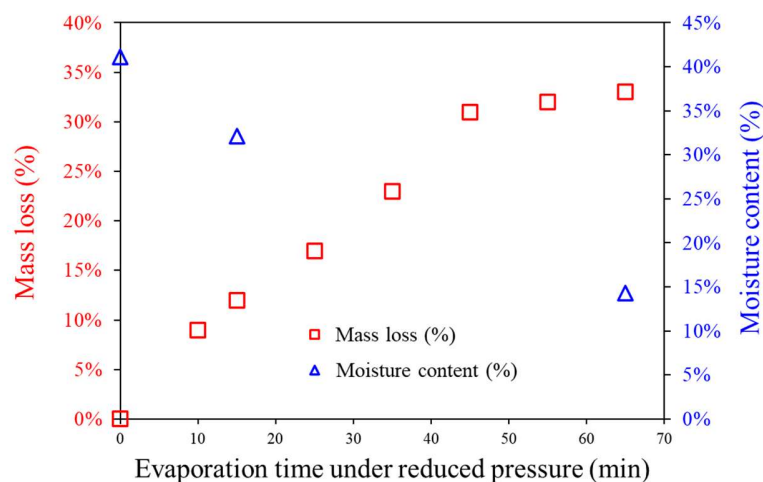


Figure 4 Mass loss (%) of experimental formulation (left axis, red color square), moisture content (%) using the Karl Fischer reagent (right axis, blue color triangle)

A rotational rheometer was used to measure how the texture of the experimental formulation changed when the phase was separated by rubbing. Strain amplitude sweep tests were performed with a rotational rheometer to measure changes in the complex modulus according to the mass loss by evaporation of the formulation. It was confirmed that the complex modulus in the phase separated formulation (Figure 5, red solid line) was reduced to less than half in the linear viscoelastic range. This suggests that the structure of the formulation has collapsed.

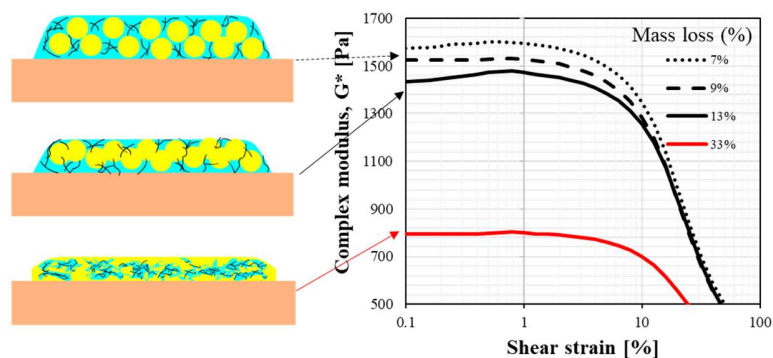


Figure 5 Strain amplitude sweep test of experimental formulation

The reason why phase transition becomes easier with the experimental formulation was analyzed using DWS measurement, which is a method capable of optically analyzing the rheological behavior of particles present in viscoelastic fluid. The behavior of emulsion present in control formulation, experimental formulation, and modified experimental formulation was analyzed. A modified experimental formulation is a formulation in which a

lipophilic thickener is added to the experimental formulation. DWS can analyze the Brownian motion of particles in the formulation.

A brief interpretation of the data in Figure 6 **Mobility trends of emulsions of each formulation inferred by DWS measurement** Figure 6 is as follows. The control formulation (Figure 6, black circle) contains twice as much hydrophilic polyacrylate thickening agent as the other two formulations. Therefore, the particles are restricted in motion. The emulsion in the control formulation exhibited a particle displacement behavior that is shorter than the emulsion of the experimental formulation. The data in Figure 6 in which the autocorrelation function for the control formulation decreases slowly for lag time is the basis for the assumption that the emulsions of this formulation are restricted in motion.

And it was estimated that emulsions of modified experimental formulation with the addition of a lipophilic thickening agent would exhibit a slower particle relaxation behavior than emulsions of the experimental formulation. This is because the density and viscosity of the emulsions increased, so the speed at which emulsions move is bound to decrease. As the data in Figure 6, the relaxation speed of the emulsions in the experimental formulation is the fastest. The next fastest relaxation speed of emulsions is the modified experimental formulation. And the slowest relaxation speed is emulsions in the control formulation.

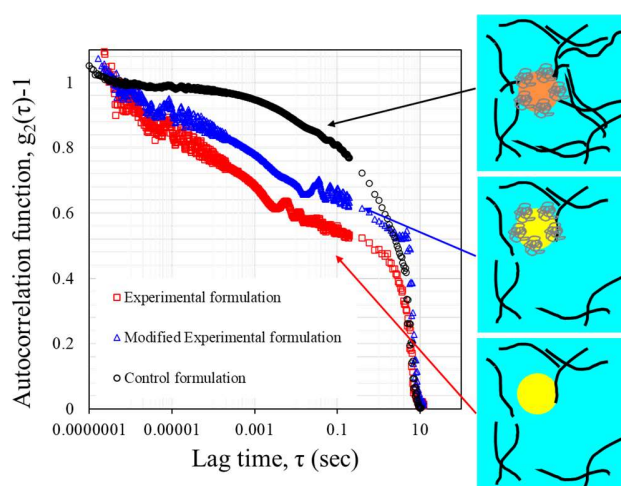


Figure 6 Mobility trends of emulsions of each formulation inferred by DWS measurement

The phase separation phenomenon of the experimental formulation by rubbing was photographed before and after rubbing with CLFM to confirm that an oil layer was indeed formed on the upper layer. Hydrophilic materials including polyol were formed on the lower layer which is close to the skin. (Figure 7) The images revealed that rubbing the experimental formulation separates the phase. And it made raised the oil layer to the upper layer.

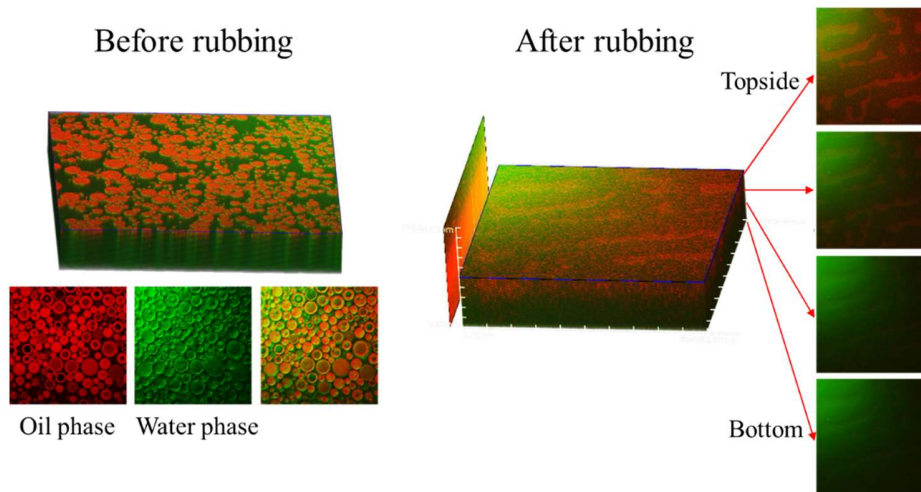


Figure 7 CLFM image of the experimental formulation. Red is fluorescence from Nile red and green is fluorescence from Nile blue.

Although the pH and salinity of the emulsion-type liquid lip formulation itself were not adjusted, Figure 8 experiments showed how pH and salinity affect skin staining in the formulation. As a result, the three kinds of acid dyes used in emulsion-type liquid lip formulation were affected by pH and NaCl concentration. Based on D&C Red No. 28, which showed the most clearly identified stain effect, the degree of staining was strong in the order of 0.1 w/v% NaCl aqueous solution > pH 3.98 aqueous solution > pH 4.7 aqueous solution > pH 8.76 aqueous solution. Likewise, other acid dyes tended to differ in staining strength depending on the pH of the aqueous solution and the presence of NaCl in the aqueous solution. An experiment was also conducted on what kind of staining tendency would appear if the acid dye was in a polyol solution. Three acid dyes were dispersed or dissolved in 1,3-butylene glycol. All samples were homogeneously dissolved or dispersed in 1,3-butylene glycol. However, there was little skin staining. Therefore, it was confirmed that for the acid dye to stain the skin, the dye must be dissolved in an aqueous solution as an ionized state.

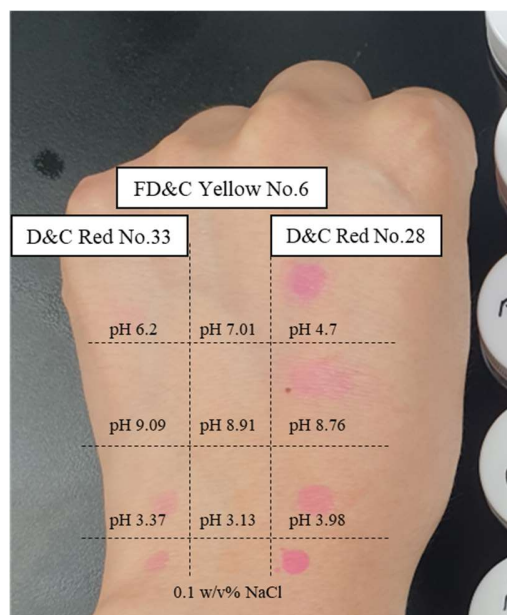


Figure 8 An experiment to determine the color staining of three acid dyes under various aqueous solution conditions.

Discussion.

In this work, we reverse the conventional notion that oil with a high refractive index of more than 1.49 should be used to show a gloss effect. We create an emulsion-type liquid lip consisting of oil with a low refractive index (1.43) with low viscosity (10.6 mPa·s). In addition, the experimental formulation also expressed a strong staining effect.

To develop such a formulation, a phase transition phenomenon by shear stress was used. Cosmetic formulations that exhibit phase transition due to shear stress have often been developed for consumers looking for unique formulations. However, efforts to identify the basic mechanism of why the phase transition phenomenon appears have been studied very little in the cosmetics industry.

In this study, various measurements and analyses were attempted to identify the phase transition phenomenon. In particular, it was meaningful to analyze the Brownian motion of emulsion particles through DWS measurements and to confirm the possibility that this could affect the coalescence of emulsion which could make phase separation phenomenon. In addition, moisture evaporates when phase separation occurs, and the method of visually checking phase separation using CLFM is a good method that can be used to simulate what happens on the skin after applying cosmetics in the future.

Based on DWS data and several other experimental data, the phase separation phenomenon of the experimental formulation can be understood as follows. When shear stress is applied to the experimental formulation, the emulsions collide with each other easily. Because the emulsions are relatively free to move compared to the emulsions in the control formulation. This can be demonstrated by the DWS measurement that the particles of the experimental formulation have a long displacement and a fast relaxation rate.

Emulsions collide by shear stress and begin to coalesce together. Unlike the control formulation, the experimental formulation, which contains only half of the water-soluble thickener, has structurally weak hindrances due to external force. And thus the O/W system is easily destroyed by shear stress. The newly discovered fact here is that the presence or

absence of a lipophilic thickening agent does not significantly affect the phase separation phenomenon caused by the shear stress of the experimental formulation. Although experimental data on this are not described in this paper, the phase separation phenomenon of modified experimental formulation easily occurred even with the addition of a lipophilic thickening agent.

In the formulation in which the emulsion structure is destroyed due to phase separation, moisture evaporates more easily. The reason is that the emulsion structure prevents water from evaporating. [12] The formulation, in which the structure of the emulsion that can prevent evaporation already disappears, partially forms a thin water film when rubbed. At this time, it is estimated that the formulation with a wider interface to the volume ratio can be easily evaporated by heat and vapor pressure. As water evaporates, water moves around polyol and water-soluble thickeners, and the density of the water phase increase rapidly, and eventually, it is completely separated into a dense water phase and a low oil phase.

The reason why the staining effect of the experimental formulation is stronger than the control formulation is estimated to be that the skin contact area of acid dyes present in the structure of the O/W emulsion is increased while the experimental formulation is phase separated by shear stress. On the other hand, in formulations that do not separate the phases even if rubbed, such as the control formulation, acid dye stays inside the structure of the emulsion.

Conclusion.

The causes and mechanisms of the two makeup effects of the experimental formulation, in which the gloss effect and the staining effect are enhanced due to the phase transition phenomenon by shear stress, were analyzed. In addition, additional experiments on the skin staining phenomenon of acid dye confirmed which factors affect it.

Based on this study, it is expected that formulators who develop liquid lip formulations will be able to receive help in the future.

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Conflict of Interest Statement. NONE.

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