

IFSCC 2025 full paper (IFSCC2025-280)

"New Standard in Skin Perfection: Polyion Complex-based Facial Makeup for Firmer and Smoother Skin"

Rina Mogaki ^{1,*}, Mariko Ohshima ¹, Tatsushi Isojima ¹, Yuichi Ikeda ¹, Kazunori Ogami ¹, Mariko Okamoto ¹, Arisa Yamana ¹, Annie Bossiere ², Thomas Pocard ², Anne Magnan ², Mai Edagawa ¹, Kumiko Uodaira ¹, Yoriko Kawaratani ¹, Ying Yin ¹

¹ L'ORÉAL R&I Japan, Kawasaki, Japan

² L'ORÉAL R&I, Chevilly-Larue, France

1. Introduction

Balancing oil control and hydration has been a long-standing challenge for cosmetics due to their conflicting nature. Especially in facial makeup, products that can achieve oil control and therefore deliver long-lasting matte are highly demanded for keeping makeup finish throughout the day. Conventional approach for oil control and matte lasting has relied on increasing oil-absorbing fillers in the formulation [1]. Such fillers may even absorb necessary sebum, resulting in dry skin. In contrast, applying oil-rich formulation is promising for skin moisturization [2]. However, excess oil often remains on the skin, which may cause an oily appearance.

To tackle this challenge, we took note of polyion complexes (PICs). In general, PICs are composed of oppositely charged polyelectrolytes which self-assemble in aqueous media via electrostatic interaction [3]. Previously, we developed a new class of PIC consisting of ionic polymers combined with a dynamic crosslinker [4–6]. Here we highlight our PIC composed of three eco-friendly ions: hyaluronic acid (HA), ϵ -poly-L-lysine (PLys), and phytic acid (PA) [5,6]. HA is a naturally occurring anionic polysaccharide, known as the major component of the extracellular matrix [7]. Due to its high water-retaining capacity, HA contributes to skin hydration and plumpness in human body [7]. HA is composed of repeating disaccharide units and forms a ribbon-like structure in aqueous solution via intramolecular hydrogen bonding [7]. PLys is a cationic antimicrobial polypeptide produced by bacterial fermentation, consisting of L-lysines linearly conjugated between the ϵ -amino and α -carboxyl groups [8]. PA, also called inositol hexaphosphate, is a natural chelating agent found in plant seeds and cereal grains [9]. Bearing six phosphate groups, PA can bind to polycations via multiple electrostatic interactions and serves as a crosslinker [10]. We reported that an aqueous mixture of PLys and PA in a specific ratio yields a dispersion of coacervate droplets (PLys-PA complex; Figure 1) through spontaneous liquid-liquid phase separation (LLPS), owing to the dynamic crosslinking of PLys by PA [5]. We also found that a PIC of HA, PLys, and PA can be obtained (Figure 1) while preventing an aggregate formation between HA and PLys by controlling their ratio and pH. We demonstrated that an aqueous skincare formulation containing this PIC forms a homogeneous, flexible film on skin and improves skin conditions [6]. The complexation is suggested to reduce the entanglement of HA, which likely forms a network in the film together

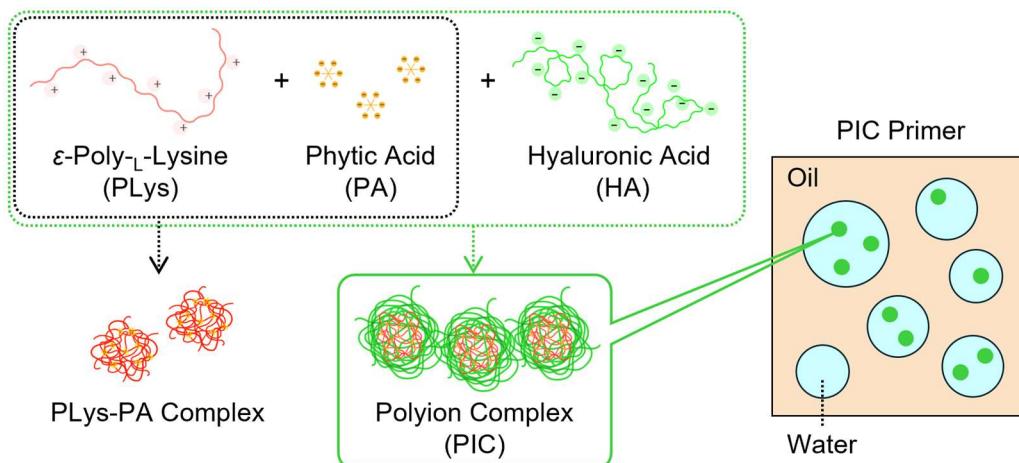


Figure 1. Schematic illustration of water-in-oil (W/O) PIC Primer containing a polyion complex (PIC) composed of hyaluronic acid (HA), ϵ -poly-L-lysine (PLys), and phytic acid (PA) in its water phase. PLys and PA forms a coacervate (PLys-PA complex).

with the condensed coacervate upon dryness. The dynamic crosslinking of the polymers most likely contributes to the flexibility of the film. These findings prompted us to utilize such PIC film as a long-lasting hydration film.

In the present work, we applied our functional PIC into a water-in-oil (W/O) makeup primer. W/O-type emulsions are advantageous in terms of water resistance particularly in makeup products. We envisioned that, if a hydration mechanism could be incorporated into a W/O formulation, balancing of oil control and hydration might be achieved in combination with the effect of fillers. However, previous reports mainly focus on coacervate systems in water [11] and not in the coexistence of oils. Moreover, a key concern in incorporating a PIC into the dispersed phase of an emulsion is whether it forms a film in a similar manner to aqueous formulations. Herein we successfully incorporated the PIC into a water phase of a W/O liquid primer originally designed to deliver oil control (PIC Primer; Figure 1) and investigated the properties of the film it forms. Despite carrying PICs in the internal water phase, PIC Primer also forms a uniform film consisting of PIC network. We demonstrate that this primer, when applied on skin, can balance oil control and hydration. Notably, PIC Primer was found to significantly improve tactile skin smoothness and elasticity in a clinical study.

2. Materials and Methods

2.1. Materials

Unless otherwise noted, cosmetic raw materials and reagents were used as received from commercial sources without further purification. 8-Hydroxypyrene-1,3,6-trisulfonic acid trisodium salt (pyranine) and Nile Red were purchased from Sigma-Aldrich.

2.2. Preparation of W/O Model Emulsion

To an aqueous solution of a mixture of sodium hyaluronate (HA; 1,100 kDa; 0.66 wt%) and PLys (4 kDa; 0.33 wt%) at pH above 10 was added phytic acid (0.18 wt%) to pH 5, followed by oleic acid (0.07 wt%). 12 g of the resultant water phase was added to an oil phase composed of *Glycine soja* (soybean) oil (100 g) and polyglycerol-6 polyricinoleate (PGPR; 6 g), and the mixture was emulsified. For fluorescence microscopic observation, pyranine and Nile Red were added to the water and oil phases, respectively.

2.3. Preparation of Primer Formulations

The compositions of the primer formulations used in this research are shown in Table 1. A water phase (Phase B; Table 1) was added to an oil phase (Phase A; Table 1), and the mixture was emulsified. Alcohol (Phase C; Table 1) was added to the resultant emulsion to obtain a liquid W/O primer formulation. The water phase of PIC Primer (Table 1) was prepared in a similar manner to that of the model emulsion. A reference primer without PLys and PA (HA Primer; Table 1) was likewise prepared using a water phase containing HA. PIC-D Primer (Table 1) with double the amount of PLys and PA compared to PIC Primer was also prepared. In addition, PIC Primer containing pyranine in its water phase with and without Nile Red in its oil phase, and HA Primer containing pyranine in its water phase were prepared for fluorescence microscopic observation.

Table 1. Compositions of PIC Primer, HA Primer, and PIC-D Primer.

Phase	Ingredients	Composition (wt%)		
		PIC Primer	HA Primer	PIC-D Primer
A	Oils	Q.S.	Q.S.	Q.S.
	Surfactants	5.00	5.00	5.00
	Fillers Pigments	11.60	11.60	11.60
	Film Formers	6.10	6.10	6.10
	Thickeners			
	Antioxidants			
B	Water	26.00	26.00	26.00
	Sodium Hyaluronate (HA) Carboxymethyl Cellulose	0.25	0.25	0.25
	ϵ -Poly-L-Lysine (PLys)	0.13	-	0.25
	Phytic Acid (PA)	0.07	-	0.14
	Oleic Acid	0.03	0.03	0.03
	Humectants Preservatives	11.70	11.70	11.70
C	Alcohol	5.00	5.00	5.00

2.4. Bright-field and Fluorescence Microscopies

Bright-field and fluorescence microscopies were performed on a Keyence model BZ-X710 all-in-one fluorescence microscope. For the observation of primer films, a microscope glass slide was spin-coated with a primer formulation containing pyranine in its water phase at 700 r.p.m. for 30 s. After drying for 3 h at room temperature, the glass slide was subjected to fluorescence microscopy ($\lambda_{\text{ex}} = 470$ nm, $\lambda_{\text{obs}} = 525$ nm). Then, an artificial sebum solution containing Nile Red was sprayed to the glass slide from a 15-cm distance. Subsequently, the glass slide was subjected to fluorescence microscopy ($\lambda_{\text{ex}} = 470$ nm, $\lambda_{\text{obs}} = 525$ nm; $\lambda_{\text{ex}} = 560$ nm, $\lambda_{\text{obs}} = 630$ nm).

2.5. Rheological Measurement

Rheological measurement was performed on a TA Instruments model Discovery HR-2 rheometer. A dried primer sample was obtained by heating a primer formulation for 2 days at 70 °C. The sample was placed under a 20-mm-diameter cone plate with a gap of 56 μm . The recovery property of the sample in response to applied shear forces at 25 °C was measured in the following programmed procedure at a constant frequency of 10 rad s^{-1} [applied shear force expressed in terms of strain (γ); duration in parentheses]: 1% (60 s), 1000% (60 s), 1% (300 s).

2.6. *In vivo Studies*

2.6.1. General

The studies were conducted in accordance with the most recent recommendations of the World Medical Association (Declaration of Helsinki), local regulations, and ICH Guideline for Good Clinical Practices. All subjects were informed about the objective of the study and provided informed consent.

2.6.2. Transepidermal Water Loss (TEWL) Measurement

TEWL measurement was performed using a Courage+Khazaka Electronic model Tewameter[®] TM 300 measuring device. The study was conducted in France on Caucasian women ($N = 12$) from 20 to 69 years old with dry skin nature on their inner forearms (initial TEWL value $\geq 6 \text{ g m}^{-2} \text{ h}^{-1}$). A primer formulation was applied to a 4-cm diameter circular area on the inner forearm with a fingerstall. One area was untreated with a primer as a control area. PIC Primer, HA Primer, and PIC-D Primer were used in this study. TEWL values were measured before application (T_0), and then 1 h, 4 h, and 8 h after application ($T_{1\text{h}}$, $T_{4\text{h}}$, and $T_{8\text{h}}$, respectively) without wiping off the formulations. Whole study was conducted in a temperature- and humidity-controlled room.

2.6.3. Facial Shine Measurement

Facial shine measurement was performed using a Bossa Nova Vision model SAMBA polarization-difference digital camera. The study was carried out in Japan on 9 Japanese women (half face each, $N = 18$, mean age 54 years old) with oily or combination skin. First, entire face was washed. Then, standard skin care products were applied to the entire face and dried. A primer formulation was applied to the entire face, followed by a powder foundation. PIC Primer, PIC-D Primer, and two primer products on the market claiming matte finish (Primers A and B) were used in this study. Whole study was conducted in a temperature- and humidity-controlled room. Polarimetric images were recorded after skin care (T_0), immediately after powder foundation application (T_{imm}), and then 3 h later ($T_{3\text{h}}$). Images under parallel- and cross-polarized lights were obtained. The shine of a given facial region was calculated using an image analysis software by subtraction of the two images, yielding a shine index in gray levels [12].

2.6.4. Clinical Study

The study was conducted in France on 59 Caucasian women in total (2 groups, $N = 28$ and $N = 31$) from 30 to 60 years old with dry skin, whose Fitzpatrick skin prototypes were from II to III. Subjects were equally divided into 2 groups based on the clinical scores of fine lines, wrinkles, skin elasticity, and skin plumpness evaluated prior to the study. The subjects were each given either PIC Primer ($N = 28$) or HA Primer ($N = 31$), together with a liquid foundation and a face cream. They were instructed to use the primer and the foundation every morning instead of their daily makeup base products for 28 days. The subjects visited the test center prior to treatment initiation (Day 0), then 7 days and 28 days after treatment initiation (Day 7 and Day 28, respectively) without product application. The face cream was applied every morning and evening from 7 days before Day 0 to Day 28. The following skin attributes that may be improved by hydration were assessed by trained experts on a scale from 0 (best) to 9 (worst) on Days 0, 7, and 28: fine line, wrinkle, skin smoothness (visual), skin texture (tactile), skin plumpness (visual), skin elasticity (tactile), and pore. Temperature and humidity were controlled during the study visits.

3. Results

3.1. Incorporation of PIC into W/O-type Emulsions

At first, we investigated the incorporation of the PIC composed of HA, PLys, and PA into a W/O-type emulsion. A water phase containing the PIC was emulsified in a mixture of *Glycine soja* (soybean) oil and PGPR. Hydrophilic and hydrophobic fluorescent dyes, pyranine and Nile Red, were added to the water and oil phases, respectively. As shown in Figure 2a, a W/O-type emulsion was obtained with a Nile Red-labeled oil as the continuous phase. A strong fluorescence emission of pyranine was observed from the inner droplet of the dispersed water phase (Figure 2a, right). Due to its anionic nature, pyranine may interact with cationic PLys in the existence of PIC. Hence, this result suggests that the water phase exists in the emulsion as a multiphase droplet containing PIC.

Next, we applied our PIC to an oil-control makeup formulation. PIC Primer was prepared by incorporating the PIC into a water phase of a W/O liquid primer formulation (Table 1). PIC Primer fluorescently labeled with pyranine and Nile Red in its water and oil phases, respectively, was subjected to fluorescence microscopy. In wet state, the continuous phase is oil, which is analogous to the model emulsion (Figure 2b, left). Interestingly, a fluorescence emission due to pyranine was observed as a continuous structure after a certain period of time (Figure 2b, right), suggesting that a phase transition occurred during the drying process. This may allow the condensation of coacervate to form a PIC network in the film upon dryness.

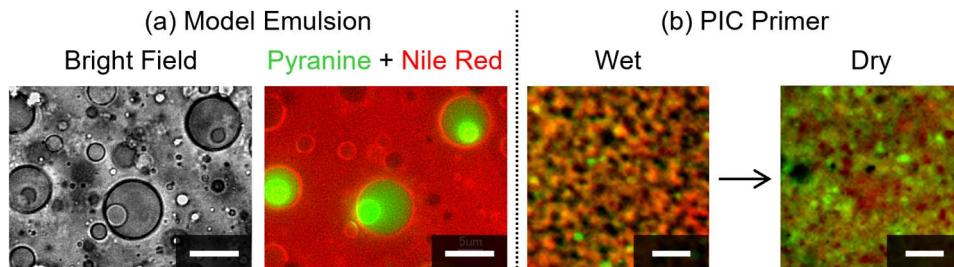


Figure 2. (a) Bright-field (left) and fluorescence (right; $\lambda_{\text{ex}} = 470 \text{ nm}$, $\lambda_{\text{obs}} = 525 \text{ nm}$, green; $\lambda_{\text{ex}} = 560 \text{ nm}$, $\lambda_{\text{obs}} = 630 \text{ nm}$, red) micrographs of W/O model emulsion containing pyranine and Nile Red. Scale bars = 10 μm . (b) Fluorescence micrographs ($\lambda_{\text{ex}} = 470 \text{ nm}$, $\lambda_{\text{obs}} = 525 \text{ nm}$, green; $\lambda_{\text{ex}} = 560 \text{ nm}$, $\lambda_{\text{obs}} = 630 \text{ nm}$, red) of PIC Primer containing pyranine and Nile Red before (left) and after (right) drying. Scale bars = 10 μm .

3.2. Properties of Primer Films

We examined the structure of the film that PIC Primer forms upon dryness. A microscopic glass slide was spin-coated with PIC Primer containing pyranine in its water phase. After drying, the glass slide was subjected to fluorescence microscopy. As shown in Figure 3a, a fluorescence emission due to pyranine was observed uniformly on the glass slide, suggesting that the PIC in the dispersed phase of the emulsion can also form a network upon drying. In sharp contrast, an uneven film with sparse non-fluorescent areas was observed for a glass slide spin-coated with HA Primer without PLys-PA complex (Figure 3b). Without the coacervate of PLys and PA, HA in the water phase exists in its entangled form, which most likely hinders the formation of a dense network upon dryness.

To further confirm the network structure of the film, we examined the recovery property of the primer films. Dried samples of PIC Primer and HA Primer, obtained by heating each formulation, were subjected to rheological measurements with an alternate step strain (Figures 3c and 3d). The initial storage modulus (G') of PIC Primer and HA Primer at a strain of $\gamma = 1\%$ were 8.1 kPa (Figure 3c) and 15.6 kPa (Figure 3d), respectively. PIC Primer gave a lower G'

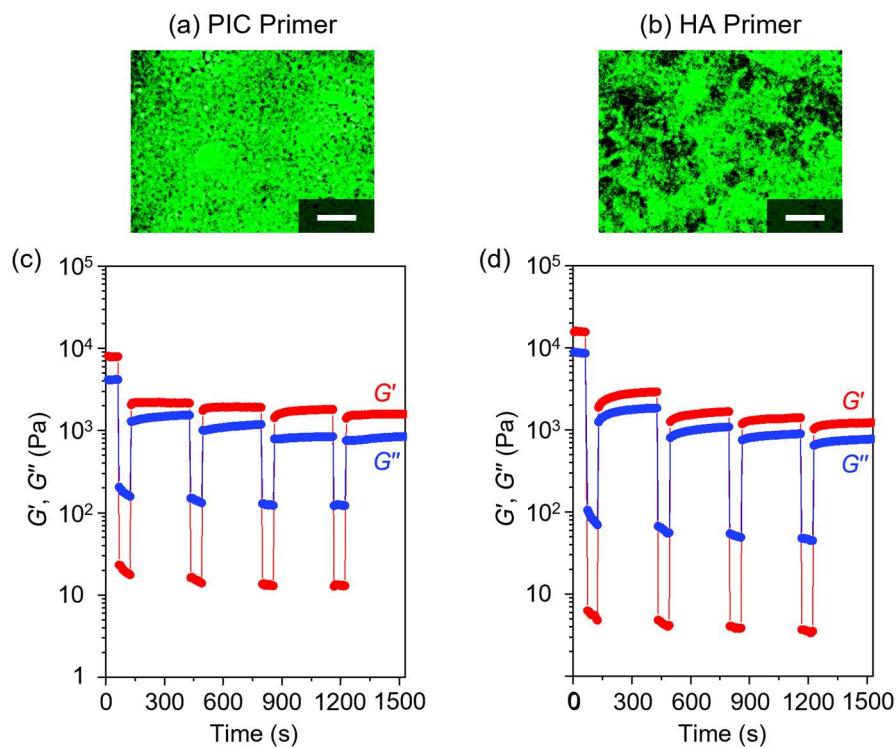


Figure 3. (a, b) Fluorescence micrographs ($\lambda_{\text{ex}} = 470 \text{ nm}$, $\lambda_{\text{obs}} = 525 \text{ nm}$) of microscopic glass slides spin-coated with (a) PIC Primer and (b) HA Primer containing pyranine. Scale bars = $20 \mu\text{m}$. (c, d) Storage (G' ; red) and loss modulus (G'' ; blue) values of dried (c) PIC Primer and (d) HA Primer in alternate step strain ($\gamma = 1\%$ and 1000% ; 10 rad s^{-1}) measurements.

value than HA Primer, indicating that PIC Primer forms a softer film. Under the application of a large-amplitude oscillatory force ($\gamma = 1000\%$; frequency 10 rad s^{-1}), G' value of PIC Primer decreased and became smaller than the loss moduli (G'' ; Figure 3c). However, when the amplitude was decreased ($\gamma = 1\%$) at the same frequency, G' value immediately recovered. The recovery rate of the film mechanical property was calculated as the ratio of the log G' values before and after four strain cycles. The recovery rate of PIC Primer was obtained as 82%, which was larger than that determined for HA Primer (74%; Figure 3d). Namely, PIC Primer film exhibits self-recovery property, again suggesting the existence of a flexible PIC network.

We then investigated the stability of the film against sebum. The microscopic glass slide spin-coated with a primer formulation containing pyranine was sprayed with an artificial sebum solution containing Nile Red. In the case of the film of HA Primer without PLys-PA complex, the fluorescence emission of pyranine (Figure 4b, green) diminished around the sebum

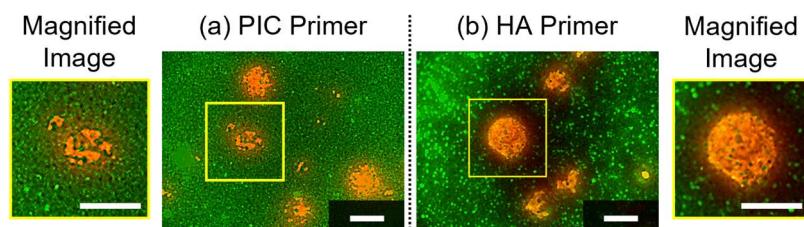


Figure 4. Fluorescence micrographs of microscopic glass slides spin-coated with (a) PIC Primer and (b) HA Primer containing pyranine recorded after spraying an artificial sebum solution ($\lambda_{\text{ex}} = 470 \text{ nm}$, $\lambda_{\text{obs}} = 525 \text{ nm}$, green; $\lambda_{\text{ex}} = 560 \text{ nm}$, $\lambda_{\text{obs}} = 630 \text{ nm}$, red). Scale bars = $100 \mu\text{m}$.

droplets (Figure 4b, red), probably due to the deterioration of the film by the sebum. Notably, PIC film maintained its homogeneous film even after sebum application (Figure 4a).

3.3. Hydration Property of PIC Primer: TEWL Suppression

The above observations suggesting the formation of a uniformly networked film of PIC Primer prompted us to investigate if the film can contribute to skin hydration. The skin barrier efficacy of the primer formulations was evaluated on human forearms ($N = 12$) by measuring the TEWL before (T_0) and 1 h, 4 h, and 8 h after application (T_{1h} , T_{4h} , and T_{8h} , respectively). Compared to the control area untreated with the formulations (Figure 5, gray), the suppression of TEWL for the area treated with PIC Primer was significant at T_{1h} ($p < .05$, Student's t -test) and exhibited tendencies ($p < .10$) up to T_{8h} (Figure 5, green). In contrast, HA Primer without PLys-PA complex significantly suppressed TEWL at T_{1h} and exhibited tendency at T_{4h} but not at T_{8h} (Figure 5, orange). Interestingly, PIC-D Primer with double the amount of PLys-PA complex significantly suppressed TEWL until T_{8h} (Figure 5, blue) compared with the control area. These results suggest that PIC Primer exhibits skin barrier efficacy owing to the PIC in the water phase. The amount of PLys-PA complex in the formulation may possibly affect the density of the PIC network, resulting in longer skin barrier effect.

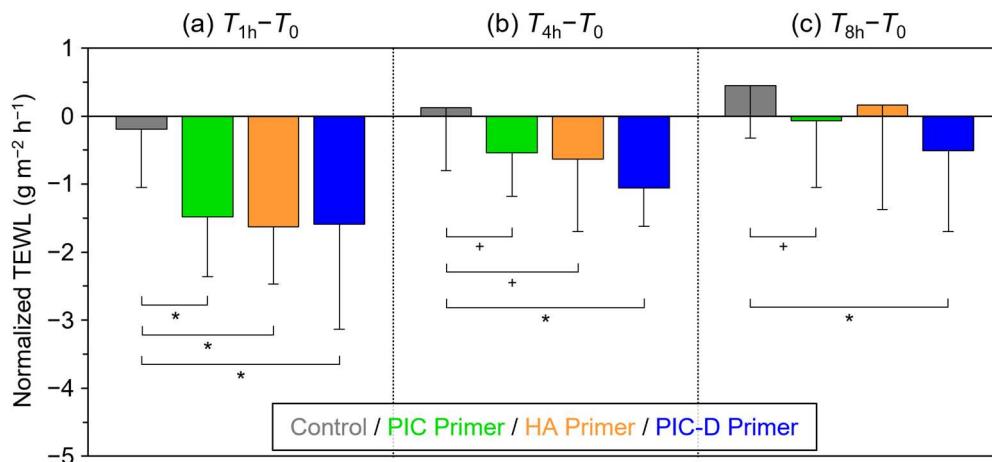


Figure 5. Normalized TEWL values ($N = 12$) at (a) T_{1h} , (b) T_{4h} , and (c) T_{8h} of a control area (gray) and areas treated with PIC Primer (green), HA Primer (orange), and PIC-D Primer (blue). The values represent the means – SD (+ $p < .10$, * $p < .05$, Student's t -test).

3.4. Oil Control Property of PIC Primer: Matte Lasting

We next investigated the oil control property of PIC Primer. As described earlier, PIC Primer was developed by incorporating PIC into a water phase of a W/O primer formulation originally designed to deliver oil control. The matte lasting efficacy of the primer formulations was evaluated on human faces ($N = 18$) by calculating the facial shine using polarimetric images recorded after skin care (T_0), immediately after powder foundation application (T_{imm}), and then 3 h later (T_{3h}). Compared to the matte-finish products on the market (Primers A and B; Figure 6a, pink and purple, respectively), the shine values were significantly lower ($p < .05$, one-sample t -test) with the faces applied with PIC Primer (Figure 6a, green) or PIC-D Primer (Figure 6a, blue). The matte lasting efficacy was evaluated by the increase of shine values at T_{3h} from T_{imm} (Figure 6b). PIC Primer and PIC-D Primer exhibited significantly higher matte lasting efficacy compared to Primer A ($p < .05$, ANOVA with Tukey's multiple comparison test) and comparable efficacy to Primer B ($p > .10$). The amount of PLys-PA complex in PIC Primer and PIC-D Primer did not affect their matte effects at T_{imm} and T_{3h} as well as matte lasting

efficacies. These results, together with the skin barrier efficacy demonstrated by TEWL suppression, allows us to conclude that PIC Primer can balance both oil control and hydration.

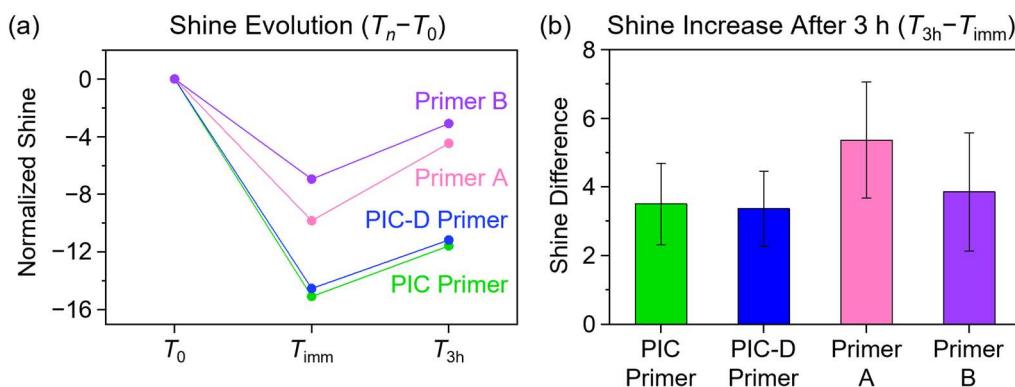


Figure 6. (a) Normalized shine values ($N = 18$) of the faces treated with PIC Primer (green), PIC-D Primer (blue), Primer A (pink), and Primer B (purple) at T_0 , T_{imm} , and T_{3h} . (b) Differences of the shine values ($N = 18$) at T_{3h} from T_{imm} of the faces treated with PIC Primer (green), PIC-D Primer (blue), Primer A (pink), and Primer B (purple). The values represent the means $\pm 95\%$ CI.

3.5. Clinical Study

Of particular interest, PIC Primer significantly improves skin smoothness and elasticity in the long term. We conducted a clinical study to explore the effect of PIC Primer achieving both oil control and hydration on skin conditions. The subjects were instructed to use either PIC Primer ($N = 28$) or HA Primer ($N = 31$) for 28 days. Skin attributes were assessed by trained experts on a scale from 0 (best) to 9 (worst) prior to treatment initiation (Day 0), then 7 days and 28 days after treatment initiation (Day 7 and Day 28, respectively). Among the attributes evaluated, the clinical score of “skin texture (tactile)”, assignable to skin smoothness, significantly improved for PIC Primer users at Days 7 and 28 compared to that of Day 0 ($p < .05$, ANOVA followed by Dunnett test; Figure 7a, green). PIC Primer exhibited the effect earlier than HA Primer which showed significant improvement only at Day 28 (Figure 7a, orange). Moreover, the improvement rate at Day 28 from Day 0 (Figure 7b) was significantly higher for PIC Primer (23.5%) compared with HA Primer (11.0%; $p < .05$, Kruskal-Wallis test). The clinical score of “skin elasticity (tactile)” significantly improved for both PIC Primer and HA Primer users at Days 7 and 28 (Figure 7c). Noteworthy, the improvement rate at Day 28 (Figure 7d) of PIC Primer (30.2%) was significantly higher than that of HA Primer (17.3%). Furthermore, both PIC Primer and HA Primer significantly improved the score of “skin smoothness (visual)” at Day 28, while PIC Primer exhibited a trend ($p < .10$) towards higher improvement rate than HA Primer. In the case of “skin plumpness (visual)”, HA Primer significantly improved the scores at both Days 7 and 28, whereas PIC Primer users only exhibited significant improvement at Day 28. However, there was no significant difference in improvement rate between PIC Primer and HA Primer at Day 28. No significant change was observed for other attributes such as “fine line”, “wrinkle”, and “pore”.

4. Discussion

Herein, we demonstrated that PIC Primer, carrying a functional PIC composed of HA, PLys, and PA in its water phase, forms a uniform film consisting of PIC network. Microscopic observation of the model emulsion in *Glycine soja* (soybean) oil suggest that the dispersed water phase exists in the emulsion as a multiphase droplet containing a condensed phase of PIC formed by LLPS. During the drying process, PIC Primer undergoes a phase transition,

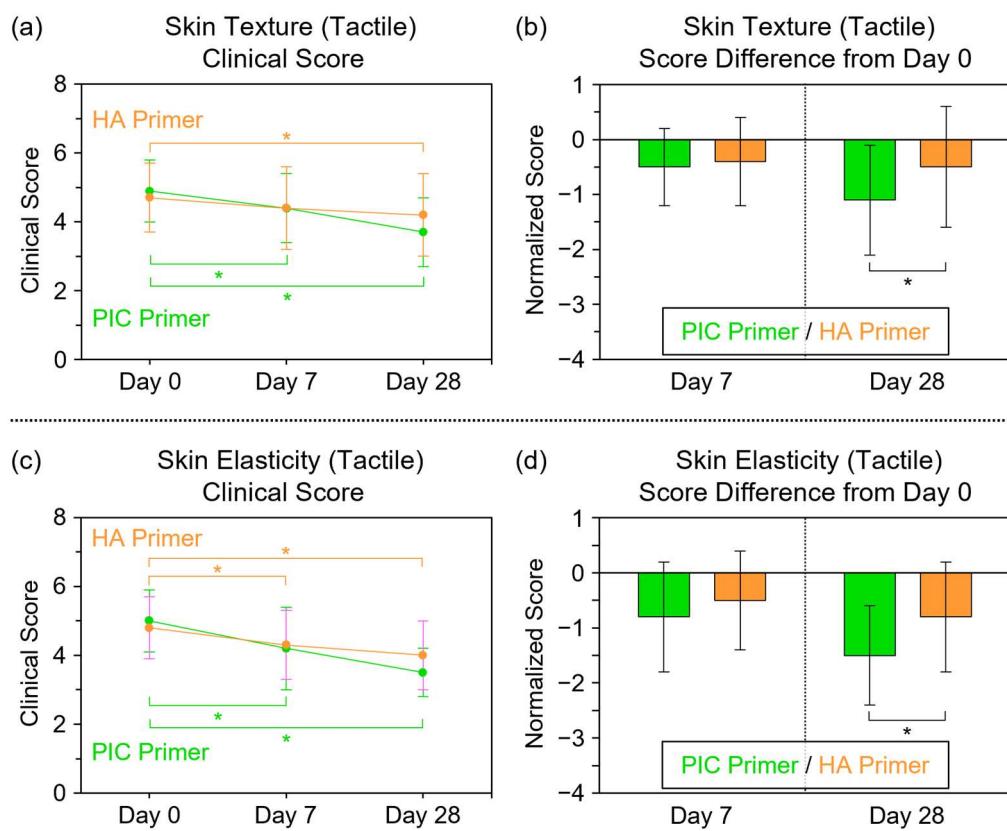


Figure 7. (a, c) Clinical scores of (a) skin texture (tactile) and (c) skin elasticity (tactile) at Days 0, 7, and 28 of the subjects who used PIC Primer (green; $N = 28$) and HA Primer (orange; $N = 31$). The values represent the means \pm SD ($* p < .05$, ANOVA followed by Dunnett test). (b, d) Normalized clinical scores of (b) skin texture (tactile) and (d) skin elasticity (tactile) at Days 7 and 28 of PIC Primer (green; $N = 28$) and HA Primer (orange; $N = 31$) users. The values represent the means \pm SD ($* p < .05$, Kruskal-Wallis test).

most likely due to the evaporation of volatile oils in the formulation. Considering the surface tension, the remaining oils likely segregate to the air interface, while the water phase condense on the substrate surface. Consequently, a network of condensed coacervate together with the detangled HA possibly forms in the film upon dryness, in spite of the fact that the PIC is initially contained in the dispersed phase of the emulsion. Owing to this complex network structure, the film likely exhibits self-recovery property as shown in rheological measurements. Moreover, the stability of the PIC film against artificial sebum suggests the possibility that the film also serves to grip the fillers in the oil phase on the film surface, which may result in efficient sebum absorption. In sharp contrast, the uneven film of HA Primer without PLys-PA complex may lose its hydrophilic-hydrophobic balance upon contact with artificial sebum, leading to deterioration.

We achieved in balancing oil control and hydration *in vivo* with our PIC Primer. TEWL measurement revealed that PIC Primer exhibits skin barrier efficacy longer than HA Primer. As suggested from *in vitro* investigation, the film of HA Primer likely deteriorates upon contact with sebum. Since sebum is continuously secreted from the skin, such film defects may allow water loss through the epidermis. In the case of PIC Primer, we consider that its film lasts longer on skin due to its stability against sebum and self-recovery property. This is also advantageous for achieving matte lasting by maintaining oil-absorbing fillers on the film surface, as demonstrated in facial shine measurements.

In the clinical study, we found that PIC Primer significantly improves tactile skin smoothness and elasticity in a long term. Dry skin lacking sufficient water and sebum is known to cause

skin roughness and a loss of stratum corneum elasticity [13]. We hypothesize that daily usage of PIC Primer can bring an optimal oil-water balance to the skin, leading to a healthier skin. Here we did not observe improvements in the attributes related to wrinkles and pores. Since these attributes are related to skin elasticity, we may possibly observe their improvements in a longer usage. We demonstrated in this work that our PIC can be incorporated into emulsions, broadening its application to various cosmetic formulations. The effect of our PIC on skin conditions shows great potential and therefore is worthy of future investigation.

5. Conclusion

In conclusion, we demonstrated that W/O-type PIC Primer, containing a PIC composed of eco-friendly ions HA, PLys, and PA in its water phase, can balance oil control and hydration. PIC Primer forms a uniform film composed of condensed PIC network, which allows to incorporate hydration mechanism into an oil-control formulation. Notably, long term usage of PIC Primer significantly improves skin smoothness and elasticity, confirmed in a clinical study. Our novel approach not only delivers a daylong matte finish but also offers an innovative concept of skin improvement through everyday makeup.

6. References

- [1] Nomura K (2000) Technique for Long-lasting Make-up Products by Functional Materials. *J Jpn Soc Colour Mater* 73(5):255–260.
- [2] Okamoto T (2016) Formulation Strategies for Skincare Cosmetics with a High Moisturizing Effect. *J Soc Cosmet Chem Jpn* 50(3):187–193.
- [3] Insua I, Wilkinson A, Fernandez-Trillo F (2016) Polyion complex (PIC) particles: Preparation and biomedical applications. *Eur Polym J* 81:198–215.
- [4] Suga T, Isojima T, Kasai T, Shiroya T, Takahashi N, Yokoyama E, Nicolas A, Asanuma H, Koike T (2023) Polyion Complex Technology for Cosmetic Transfer Prevention. *Oleoscience* 23(1):5–10.
- [5] Isojima T, et al (2023) An investigation into the application of polyion complex containing hyaluronic acid in cosmetics. 10th Nagoya Biomimetics International Symposium, November 17, Nagoya, Japan.
- [6] Shimatani M, et al (2024) Natural and Effective Solution for Long-Lasting Hydration and Plumpness of the Skin Using Core-Shell Ion Complex. 34th IFSCC Congress, October 14–17, Iguazu Falls, Brazil.
- [7] Papakonstantinou E, Roth M, Karakiulakis G (2012) Hyaluronic acid: A key molecule in skin aging. *Derm-Endocrinol* 4(3):253–258.
- [8] Wang L, Zhang C, Zhang J, Rao Z, Xu X, Mao Z, Chen X (2021) Epsilon-poly-L-lysine: Recent Advances in Biomanufacturing and Applications. *Front Bioeng Biotechnol* 9:748976.
- [9] Silva EO, Bracarense APFRL (2016) Phytic Acid: From Antinutritional to Multiple Protection Factor of Organic Systems. *J Food Sci* 81(6):R1357–R1362.
- [10] Xu L, Wang C, Cui Y, Li A, Qiao Y, Qiu D (2019) Conjoined-network rendered stiff and tough hydrogels from biogenic molecules. *Sci Adv* 5(2):eaau3442.
- [11] Rumyantsev AM, Jackson NE, de Pablo JJ (2021) Polyelectrolyte Complex Coacervates: Recent Developments and New Frontiers. *Annu Rev Condens Matter Phys* 12:155–176.
- [12] Cassin G, Diridollou S, Flament F, Adam AS, Pierre P, Colomb L, Morancais JL, Qiu H (2018) Concealing a shiny facial skin appearance by an Aerogel-based formula. *In vitro* and *in vivo* studies. *Int J Cosmet Sci* 40:58–66.
- [13] Takahashi Y (2018) Moisturizing Cosmetics and Their Effects. *J Jpn Cosmet Sci Soc* 42(4):280–287.