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## ***Long-Lasting Skin Radiance Achieved by Hydrogel-in-Oil Emulsion***

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### **1. Introduction**

Recent researches have established a significant link between facial skin radiance and perceived attractiveness, with radiant skin contributing to a more positive overall impression [1–2]. Consequently, skin radiance is now a primary target for cosmetic product development.

Skin radiance appears to result from a harmonious combination of skin colors, light reflection, and skin texture [3]. However, numerous biological and optical characteristics of skin still influence its appearance [4]. When light penetrates the skin, it interacts with various components, such as collagen fibrils, melanin and blood vessels, causing it to scatter in different directions and then emerges, creating a soft, translucent glow. This effect is often associated with healthy and youthful skin. With skin aging, subsurface scattering tends to decrease, potentially leading to a loss of radiance and translucence.

In addition, the measurement of gloss can be influenced by the skin tone as for instance darker skin tends to scatter less light. Some glossmeters can minimize this effect by applying diffuse scattering correction, ensuring accurate gloss measurements regardless of skin color.

Current strategies for enhancing skin radiance focus on two main areas: active ingredient innovation and formulation development. Studies indicate that several active ingredients, including peptides, vitamin C, bakuchiol, and vanilla tahitensis extract demonstrate efficacy in improving skin radiance through various biological mechanisms [5–6].

In addition to active ingredients, formulation approaches also play a crucial role. The use of liquid emollients with high refractive index (RI) has been shown to enhance skin radiance [7]. Furthermore, research suggests that moisturizer formulations incorporating liquid crystal can improve skin hydration and, consequently, enhance perceived skin radiance [8].

There are various types of emulsions used in cosmetics, each with their own benefits, making them suitable for specific product applications. For enhancing skin radiance, water-in-oil (W/O) emulsions are considered to have certain advantages over oil-in-water (O/W) emulsions. This is because W/O emulsions typically contain a higher concentration of oils, and the oils in the external phase generally have a higher refractive index (RI) than water.

Given this context, this study aims to investigate how different emulsion types affect skin radiance, rather than focusing on individual ingredients, to gain deeper understanding of their impact. By using minimal numbers of ingredients, O/W emulsion and two different W/O emulsions were prepared. Skin benefits such as skin radiance, and skin hydration were evaluated and compared.

## 2. Materials and Methods

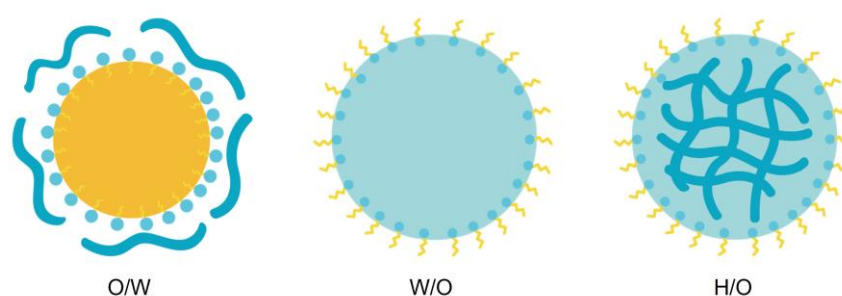
### *Preparation of emulsions*

Emulsions were prepared as described in **Table 1.**, consisting of one oil-in-water (O/W) emulsion and two water-in-oil (W/O) emulsions.

The O/W emulsion was fabricated using an O/W emulsifier (HLB 14.9) along with sodium acrylates copolymer and lecithin. The first W/O emulsion was formulated using a natural polyglyceryl fatty ester, specifically polyglyceryl-6 polyricinoleate (PG-6-PR, HLB 3.0) through direct method. The second W/O emulsion incorporated both PG-6-PR and sodium acrylates copolymer and lecithin, forming a hydrogel within the internal phase of the emulsion, resulting in a hydrogel-in-oil (H/O) emulsion.

	O/W	W/O	H/O
Water	to 100	to 100	to 100
Preservatives	3.5	3.5	3.5
Glycerin	8	8	8
O/W emulsifier (HLB: 14.9)	3		
Polyglyceryl-6 Polyricinoleate (HLB: 3.0)		5	3
Sodium acrylates copolymer and lecithin	2		2
Coco-Caprylate/Caprate	6	6	6
Squalane	6	6	6

**Table 1.** Composition of the emulsions (wt.%)



**Figure 1.** Schematic description of the prepared emulsions

### *Performance evaluation*

*In vivo* performance tests were designed to evaluate the effects of the emulsions on facial skin radiance and skin hydration in a cohort of healthy adult women. The study population comprised 10 Chinese female volunteers, aged 20 to 60 years, who met strict inclusion criteria: Fitzpatrick skin type II - III (light to moderate skin tone), normal or combination skin excluding dry or oil skin, and general good health and mental state. All subjects were required to present with a cleansed face, free from lotion, sunscreen, makeup, perfumes, or any skincare products

on the day of each visit, and to have abstained from participation in other facial studies during the trial period.

The investigational product was self-applied by each volunteer, using their fingers to tap and blend 0.6g evenly across the entire face. The evaluation protocol included multiple time points: baseline ( $T_0$ ), 30 minutes post-application ( $T_{30\text{min}}$ ), 4 hours post-application ( $T_{4\text{hrs}}$ ), and 7 hours post-application ( $T_{7\text{hrs}}$ ).

Skin glow was assessed by Skin-Glossometer® GL200 (Courage+Khazaka electronic GmbH, Köln, Germany) and skin hydration evaluation has been performed with Corneometer CM825® (Courage+Khazaka electronic GmbH, Köln, Germany). VISIA-7 imaging system device (Canfield Scientific, Parsippany, NJ, USA) was used to obtain facial images under STD light mode.

### *Electron microscopic observation*

#### *cryo-FESEM*

Zeiss Crossbeam 550 (Carl Zeiss Microscopy, Oberkochen, Germany) equipped with Quorum PP3010T cryo-SEM preparation system (Quorum Technologies, Laughton, England) was used to investigate the microstructure of the emulsions. A drop of the emulsion was mounted on aluminum specimen holder and frozen in the slushing station with liquid nitrogen. The sample was transferred to the preparation chamber under vacuum. The frozen sample was freeze-fractured at  $-175^\circ\text{C}$  and coated with platinum, then inserted into the observation chamber. The sample was examined at an accelerating voltage of 1.5 kV.

#### *cryo-FIB-SEM*

FIB-SEM Scios 2 (Thermo Fisher Scientific, Waltham, MA, USA) equipped with Quorum PP3010T cryo-SEM preparation system (Quorum Technologies, Laughton, England) was used to characterize the microstructure of the emulsions. A drop of the emulsion was mounted on aluminum specimen holder and frozen at  $-175^\circ\text{C}$  in the slushing station with liquid nitrogen. The sample was transferred to the preparation chamber under vacuum and coated with platinum. The sample was introduced into the FIB-SEM under vacuum, and cross-sectioned with FIB.

#### *cryo-FF-SEM*

FIB-SEM Scios 2 (Thermo Fisher Scientific, Waltham, MA, USA) equipped with Quorum PP3010T cryo-SEM preparation system (Quorum Technologies, Laughton, England) was used. A drop of the emulsion was mounted on aluminum specimen holder and frozen at  $-175^\circ\text{C}$  in the slushing station with liquid nitrogen. The sample was transferred to the preparation chamber under vacuum and cut with a knife. Sublimation ( $-100^\circ\text{C}$ , 10 minutes) and platinum coating were followed.

## **3. Results**

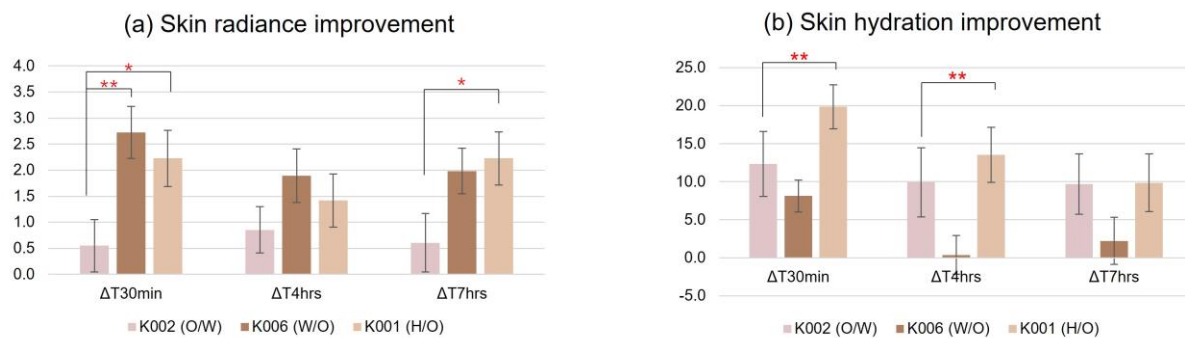
### *Skin radiance*

Glossometer measurements indicated that both the W/O emulsion and H/O emulsion showed significant improvement of skin glow at 30 minutes, 4 hours and 7 hours after application (**Figure 2. (a)**). The W/O emulsion demonstrated greater improvement up to 4 hours, after which the H/O emulsion exhibited superior radiance at the 7-hour mark. Among the emulsions, only the H/O emulsion showed a significantly improvement in radiance after 7 hours. In contrast, O/W emulsion did not demonstrate any skin glow effect at any of the measured time points (**Table 2.**).

### Skin hydration

The H/O emulsion resulted in greater improvement in skin hydration compared to the O/W and W/O emulsions (**Figure 2. (b)**). It showed the highest increase in 30 minutes, indicating that the H/O emulsion provides the greatest short-term improvement in skin hydration. While the H/O emulsion maintained enhanced skin hydration up to 7 hours, the O/W emulsion did not show a sustained effect. The W/O emulsion was found to provide only a short-term improvement (**Table 2**).

**Figure 3.** exhibits VISIA 7 images of the volunteers taken at baseline and after 7 hours. Improvement in skin radiance on the facial skin was visually evident under light, and the results showed that the H/O emulsion helped maintain a glow even 7 hours after application.



**Figure 2.** Skin radiance (a) and skin hydration (b) improvement in 30 minutes, 4 hours and 7 hours compared to the baseline, “\*” for  $p < 0.05$ , “\*\*” for  $p < 0.01$ , “\*\*\*” for  $p < 0.001$ .

Measurement	Emulsion type	T <sub>30min</sub> Vs. T <sub>0</sub>	T <sub>4hrs</sub> Vs. T <sub>0</sub>	T <sub>7hrs</sub> Vs. T <sub>0</sub>
Skin radiance	O/W	NS	NS	NS
	W/O	+27.20%***	+18.92%**	+19.83%**
	H/O	+22.66%**	+14.43%*	+22.66%**
Skin hydration	O/W	+20.35%*	NS	+15.97%*
	W/O	+12.65%**	NS	NS
	H/O	+33.92%***	+23.11%**	+16.85%*

**Table 2.** % increase of skin radiance and skin hydration, “\*” for  $p < 0.05$ , “\*\*” for  $p < 0.01$ , “\*\*\*” for  $p < 0.001$ .



**Figure 3.** Representative images taken by a VISIA 7 subject at baseline (T<sub>0</sub>) and at 7 hours (T<sub>7hrs</sub>).

## 4. Discussion

### *Skin radiance*

It was found that the two W/O-type emulsions provided improvements in skin radiance, both in the short and long term. Two main factors likely contributed to the enhanced skin glow: the emulsion type and the emulsifier, PG-6-PR.

Because the oil phase is present on the exterior of the emulsion, the oil component can effectively cover and wrap the skin surface (**Figure 4. (b),(c)**), resulting in greater light reflection and skin glow compared to the O/W emulsion.

Additionally, the lipophilic emulsifier may also play a role in these results. PG-6-PR is a thick liquid emulsifier with a naturally high refractive index. The refractive index of polyglyceryl polyricinoleate (PGPR) is typically between 1.463 and 1.468 at 20°C. Beyond its primary function as an emulsifier, the presence of 5% and 3% of the PG-6-PR in the emulsions may also contribute to increased skin radiance.

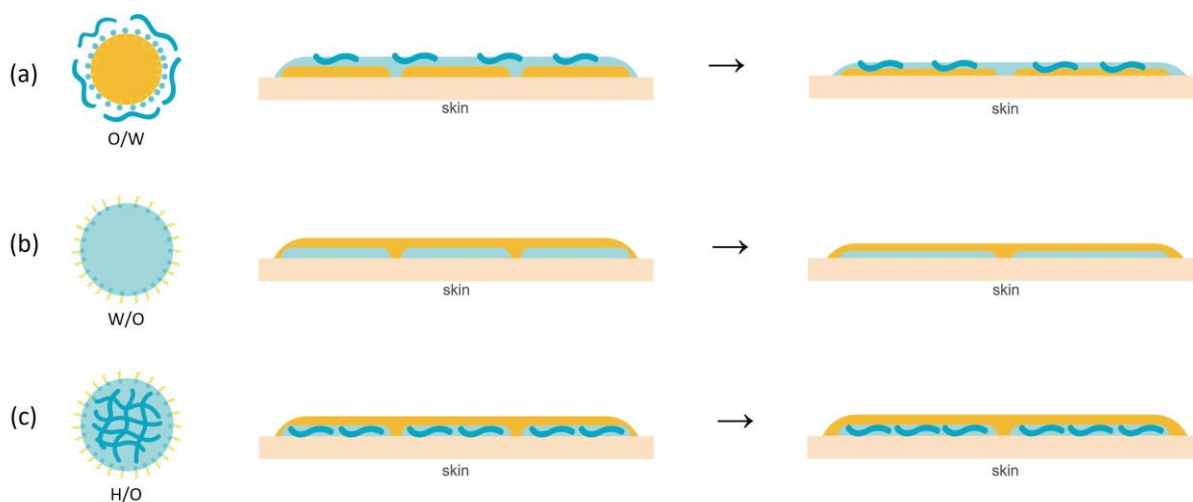
Most importantly, the hydrogel within the oil phase contributed to longer-lasting radiance compared to the traditional W/O emulsion. It is believed that the polymeric network in water phase is preserved beneath the oil phase, maintaining a firm oil residue on the skin surface (**Figure 4. (c)**).

### *Skin hydration*

The skin hydration test results revealed which ingredients and emulsion types are most effective not only for immediate hydration but also for maintaining long-lasting hydration. Skin hydration was particularly influenced by the polymer ingredient, acrylates copolymer. The polymer played a significant role in both O/W and H/O emulsions, where it left a polymeric residue on the skin. The polymer network helps retain water on the skin for a longer period, potentially delaying water evaporation after application. Slower water evaporation contributes to improved and more persistent skin hydration.

Meanwhile, the emulsion type had an even greater effect on sustained skin hydration. Although the W/O emulsion showed less improvement in hydration compared to the O/W emulsion, the H/O emulsion demonstrated higher and more continuous enhancement of skin hydration. This indicates that both the water thickener and the emulsion type are crucial for effective skin hydration.

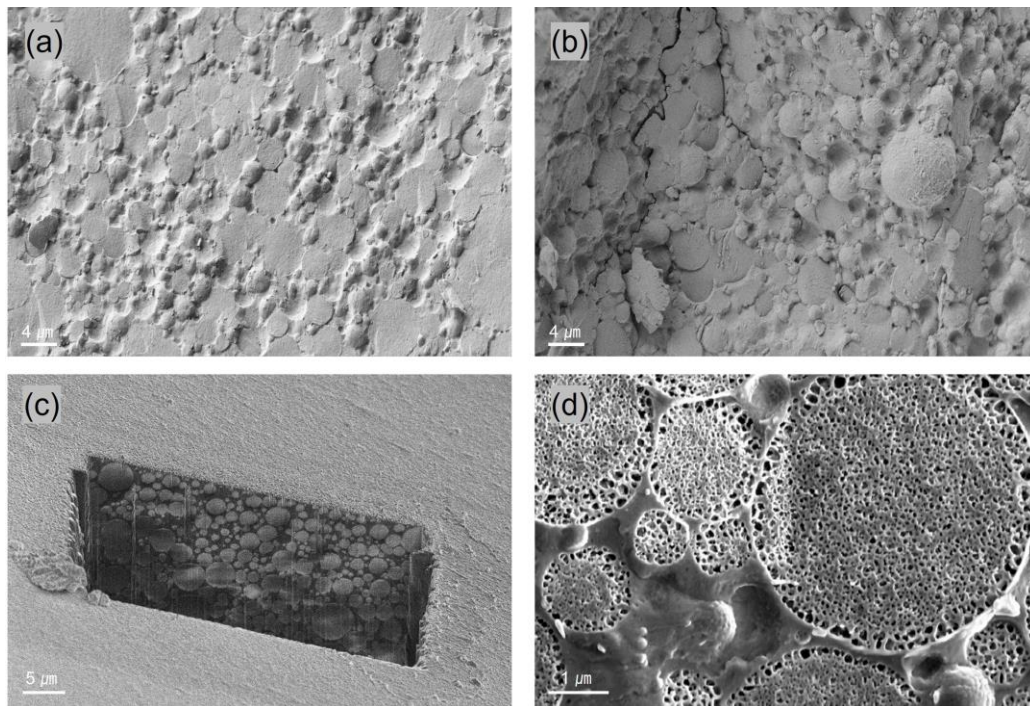
Structurally, O/W emulsions are more susceptible to water evaporation (**Figure 4. (a)**). Even though they contain a polymer network in the water phase, water can easily evaporate, leaving the polymer residue on the skin.



**Figure 4.** Different processes and results of application on the skin.

#### *Characterization of the H/O emulsion*

To investigate the mechanism of the different skin benefits of the H/O emulsion, electron microscopic observations have characterized the structure of the emulsion. The *cryo*-FESEM images (**Figure 5.** (a), (b)) show that the emulsion consists of irregular sizes of droplets packed in smaller portion of external phase. When measured by *cryo*-FIB-SEM (**Figure 5.** (c)), the emulsion showed the same configuration in the freezed cross-section. After sublimation, it was confirmed that the emulsion particles have distinct structure of internal phase, surface and external phase (**Figure 5.** (d)). In the water phase it has polymeric network by acrylates copolymer, and it has different net-like surface around the hydrogel phase. Molecular differentiation between the hydrogel and surfactant layers likely arises from distinct molecular arrangements, which may contribute to the H/O emulsion's unique functional properties. While initial hypotheses suggested a liquid crystal structure, SAXS and WAXS analyses conclusively demonstrated the absence of crystalline organization in the H/O emulsion. These findings align with *cryo*-SEM observations.



**Figure 5.** Scanning electron microscopic observations of the hydrogel-in-oil emulsion by *cryo*-FESEM, 5000x (a,b), *cryo*-FIB-SEM, 7000x (c) and *cryo*-FF-SEM after sublimation, 50000x (d).

## 5. Conclusion

In this study, we investigated skin benefits such as skin radiance and skin hydration based on different emulsion types. Three types of emulsions were evaluated and compared. The *in vivo* test results showed that both the polymeric network in the internal water phase and the external oil phase of the H/O emulsion had a significant impact on long-lasting skin radiance and prolonged skin hydration. It is believed that the unique structure of the H/O emulsion contributed to these long-lasting skin benefits.

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