

## New Film-Forming Complex for All-day Lasting Youthful Look

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### Abstract

In today's world, in-office cosmetic procedures have become commonplace among beauty savvy consumers, with majority seeking treatments for undereye bags, dark circles, fine lines, and wrinkles. However, many still prefer at-home solutions with a lower cost and risk. While many skincare technologies developed have been to counter this need gap including reactive silicones, sodium silicate and other film technologies, they are prone to subpar results due to film softening, cracking or loss of adhesion over time.

To deliver immediate, dramatic, natural-looking and all-day lasting results, a film-forming complex was developed with optimal skin adhesion, using core-shell copolymer particles as adhesive fillers dispersed in a thermoplastic elastomer in volatile oils or W/O emulsion. Modulating the ratios of polymer particles and thermoplastic elastomer allowed for the formation of a film, with desired elasticity and good adhesion, upon solvent evaporation. The elasticity and soft focus of the film can be also tuned by incorporating silica and silicone elastomer.

The efficacy of this film-forming system was confirmed via in-vitro, ex-vivo and finally in-vivo use-tests where consumers reported experiencing instant eyelift or Botox-like wrinkle smoothing, tightening, compressing, and blurring effect, with good comfort, lastingness, resistance to facial movements and easy removability.

A novel film-forming complex with tunable physico-mechanical properties was engineered for an at-home solution for instant and dramatic reduction of undereye bags, and eye/glabellar/forehead wrinkles, which can pave the way for future hybrid technologies at skincare-makeup interface, to temporarily combat signs of fatigue and aging with easy application and removability.

**Keywords:** Skin-transformation; eyelift; tightening effect; wrinkles; film forming complex

## **Introduction**

In today's world, the competition to put their "best face forward" has enhanced consumer interest in improved skin appearance and they often seek instant, dramatic changes for a more youthful appearance. While invasive and minimal-to-noninvasive cosmetic procedures have become commonplace among beauty savvy consumers, many of them still prefer at-home solutions with a lower cost and risk. Most skincare solutions are targeted to anti-aging and prevention or management and correction with a long use period before improvements can be perceived. However, the improvements, if any, are far from what can be achieved with cosmetic procedures. Some of the major reported areas of concern for the majority of the consumers include undereye bags, dark circles, fine-lines, and wrinkles.

In recent years, various skincare technologies have delved into developing new technologies to tackle this challenge and provide solutions for instant gratification results approaching procedure levels. For example, reactive-silicone dispersion was reported to cure on skin and form a thin film to reduce sagging and puffing of undereye bags by altering skin mechanics for tightening or firming effects [1]. Non-reactive film-formers, e.g., silicone-resin/film-former, latex dispersions and high molecular-weight polymers, have also been effective in enhancing the Young's Modulus of skin, reducing deep wrinkle visibility and smoothing skin or undereye bags [2-6]. Besides polymeric film technologies, sodium silicate, present in many marketed products, also makes skin-tightening films, which can reduce undereye bags and wrinkles. However, all these film technologies suffer from the lack of lasting skin firmness due to film softening, film cracking or loss of adhesion to skin over time. To fill this need gap and deliver immediate, dramatic, and natural-looking results that lasting all day long, a film-forming complex with optimal skin adhesion, using core-shell copolymer particles [7] as adhesive fillers dispersed in a thermoplastic elastomer [8] either in volatile oils or in W/O emulsion. The amount of thermoplastic elastomer and core-shell particles dictates the softness or hardness of the film, respectively. Modulating their ratios in the complex allows the resulting complex to form films, with desired elasticity and good adhesion, upon solvent evaporation. The elasticity and soft focus of the film can be further tuned by incorporating silica and silicone elastomer in the formula architecture.

## **Materials and Methods**

### Formulation

The film-forming complex engine comprised of four key ingredients: adhesive particles of acrylates/isobornyl acrylates copolymer (Mexomere PBM, Noveal FR), hydrogenated styrene/butadiene copolymer thermoplastic elastomer (Ellamera Ter Set 503, Kraton Polymers), dimethicone cross polymer (Dowsil EL-8048 ID Silicone Organic Blend, Dow Corning) and hydrophobically modified silica silylate (Dowsil VM-2270 Aerogel Fine Particles, Dow Corning). For this study, the formula was prepared as a water-in-oil (W/O) emulsion, where the polymer particles were dispersed in thermoplastic and silicone elastomers in isododecane, which formed the continuous phase. Aerogel fine particles were added after the emulsification process as filler and reinforcer. The small internal aqueous phase was finally encapsulated in superabsorbent microspheres of sodium acrylates cross polymer (Aquakeep 10SH-NFC, Sumitomo Seika). Figure 1 presents the architecture of the formula.

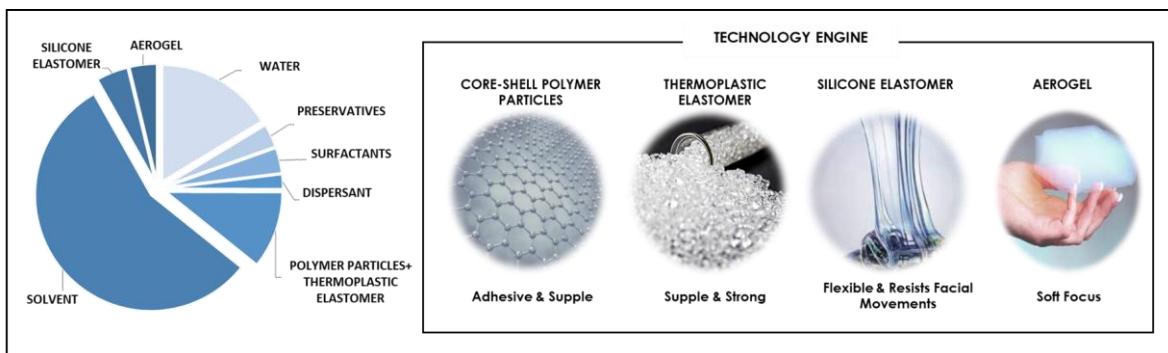


Figure 1. Architecture of the Film-Forming Complex Formula

### In Vitro evaluation

The physical chemistry of this unique film-forming complex was systematically studied through solution-rheology of the formulation. Flow profile of the formula was obtained using a TA DHR3 Rheometer. The sample was analyzed using a steel parallel plate geometry (40 mm diameter). The sample was conditioned by pre-shearing at 0.1 rad/s for 30 s,

followed by a flow ramp program with variable shear rate from 0.01/s to 1000/s for a 5 min duration, at 25°C.

*In Vitro* morphological and mechanical analyses of the film were also performed using an automated testing platform to wear-under-abrasion, tack evolution during film formation and after drying and resistance to water and sebum in accordance with the protocol detailed by Gustavo S. et al in their book chapter [9]. The film coating was prepared within the automated platform at 100 um dry thickness on a contrast card and synthetic substrate mimicking sebum and drying for 24 h at 25°C and 45% relative humidity (RH). Tack during drying was evaluated by depositing 10 µL of the sample on a contrast card. Sensitivity to water, sebum and olive oil was evaluated within the wear test parameters by depositing a 2 µL drop of water or 10 µL drop of artificial sebum or olive oil. The sensitivity of the coating to these aggressors was scored on the following scale: not solubilized, marked, tacky and solubilized.

Internal stress of the final films was analyzed by making films on a nitrile substrate and drying followed by analysis of shrinkage due to internal stress at 3 and 24 h.

For tensile test, a sample deposit was prepared by drawing down a film of 300 µm dried thickness, after drying for 7 days at 25°C and 45% RH. Measurements were performed at 3 and 24 h after drying by uniaxial tension applied on the sample on an Instron Tensile Tester at the speed of 50 mm/s, with the distance between the jaws maintained at 30mm.

Samples for the testing of optical properties of the film were prepared by drawing down the formula on transparent PET sheets using the 8 mL path of a BYK film drawdown bar. The samples were dried for 24 h under ambient conditions and the optical properties were subsequently measured using BYK haze-guard and gloss meter.

#### *Ex Vivo Evaluation*

An *ex vivo* skin model of boxcar scars was used to study the filling and lifting effect by optical coherence tomography (OCT). Additionally, film samples on ex-vivo skin were evaluated for their elasticity by dynamic mechanical analysis (DMA) and adhesion by peeling test.

#### In Vivo Evaluation

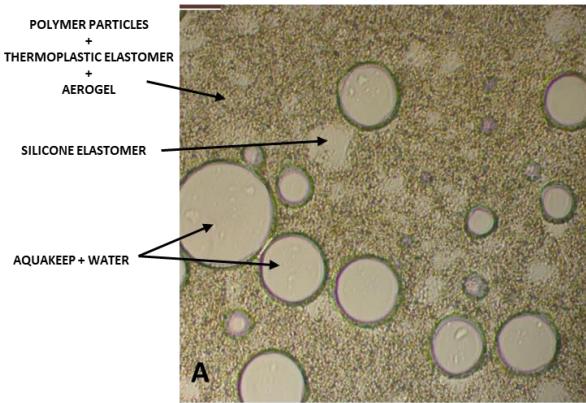
The efficacy of the film on skin under facial movement, *in vivo*, was evaluated with 3D instrumental methods by characterizing topological properties of eye bags and quantifying skin-surface strain experienced during tightening [9]. The formula was applied in a clinic setting by a skincare expert aesthetician on the panelists. Undereye bags and wrinkles were graded by the expert on the panelists in a controlled environment at before application and at 10 min, 30 min, 1 h, 3h and 6h post application, and upon removal.

The efficacy of the system was further tested and validated in an array of use-tests with over 500 consumers. The consumers were provided with the formula and asked to use it with their normal routine for up to 5 days, including a learning phase in the beginning. The consumer feedback was collected via interactive sessions and questionaries. The answers were then tabulated, and data was quantified and statistically analyzed.

## **Results and Discussion**

#### Formula Aspect

The Film-forming complex formula is a white cream with which can be easily applied with gentle force. Microscope analysis shows the formula to be a uniform dispersion with large swollen aquakeep particles with absorbed aqueous phase (Figure 2).



*Figure 2: - Micrograph of Film Forming Complex Formula (10X)*

#### Film Analysis

The coating of the formula prepared on a contrast card at 30 µm was observed to be matte and cohesive. Table 2 shows the results obtained from the automated program for these films. The film showed good wear resistance. While the film was solubilized when tested with sebum and olive oil, it showed resistance to water, although it was observed to be marked after wear test under water abrasion. There was no tack observed after drying for 24 h. The film showed moderate internal stress and had an elastic modulus of 14.8 MPa with 34% elongation at break.

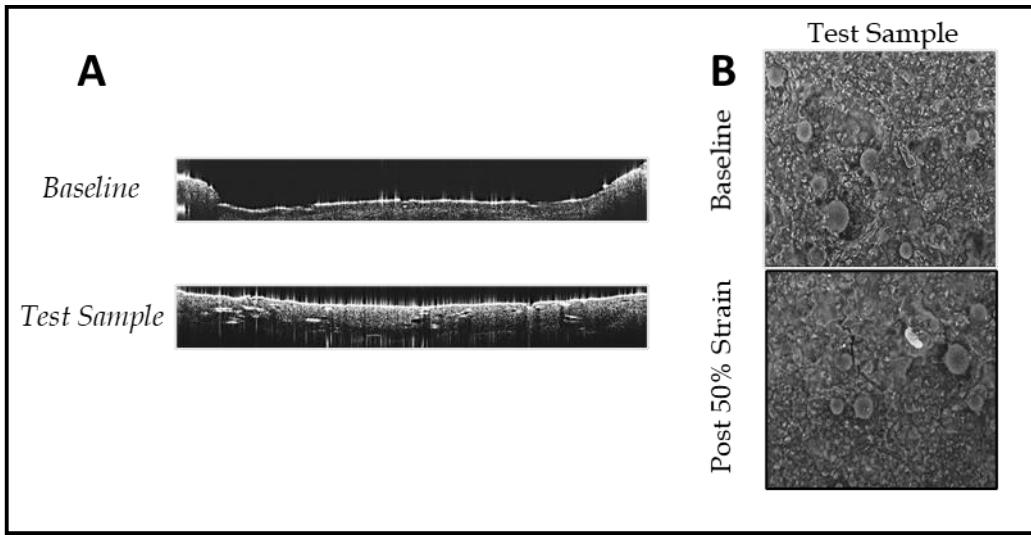
ASPECT	RESULT				
Original Film					
Wear Resistance					
	200 < strokes < 300				
Chemical resistance	<table border="1"> <tr> <td>Water (1H)</td> <td>Marked</td> </tr> <tr> <td>Sebum (24 H)</td> <td>Solubilized</td> </tr> </table>	Water (1H)	Marked	Sebum (24 H)	Solubilized
Water (1H)	Marked				
Sebum (24 H)	Solubilized				

	<b>Olive Oil (24 H)</b>	Solubilized
<b>Tack after 24 H drying on BYK contrast card and FP40</b>	0.0	
<b>Gloss at 60 °</b>	0.1	
<b>Internals Stresses at 24 H (%)</b>	10	
<b>Mechanicals</b>	<b>E (MPa)</b>	14.8
<b>Properties</b>	<b><math>\epsilon_R</math> (%)</b>	34

The optical properties of the film measured on the film drawn on a transparent sheet, showed that the films possessed transparency >90 and haze values between 90-95. These values correspond to high clarity of the film and enough haze to have a blurring effect without obstruction light. The gloss value at 60° was 8, which lies in the natural-matte zone. The Young's Modulus for the film was determined to be

#### Ex Vivo Analysis

OCT images taken of films prepared on the *ex vivo boxcar scar- skin model* showed significant lifting and filling effect (Figure 3A). The film is resilient on the skin model substrate and show durability when subjected to load to induce a 50% strain (Figure 3B). The tensile modulus and viscoelastic ratios as determined by DMA were 0.04 and 2 respectively, while the adhesion force determined by peeling test was 100 g. The results corroborate with the in vitro analysis in terms of the strength and flexibility and confirms the adhesivity of the film to skin.



*Figure 3: (A) Lifting and filling of boxcar scar model and (B) SEM image of before and after stretching the film on ex vivo skin model*

#### In Vivo Performance- Expert and Instrumental

The *in vivo* results from studies performed in controlled setting with skin care expert aestheticians showed an instant and dramatic improvement in skin tightness and texture which correlated well and was consistent with in-vitro and ex-vivo observations. The experts observed a 1-2 grade improvement in undereye bags and wrinkles and 1-1.5 grade reduction of forehead and glabellar lines (evaluated based on skin aging atlas by experts), within 30 minutes and was maintained for the duration of the study at 6h. Additionally, instrumental 3D analysis showed a 60% reduction in undereye wrinkles, 55% reduction in glabellar wrinkles and 32% reduction in depth of forehead wrinkles (Figure 4-5)

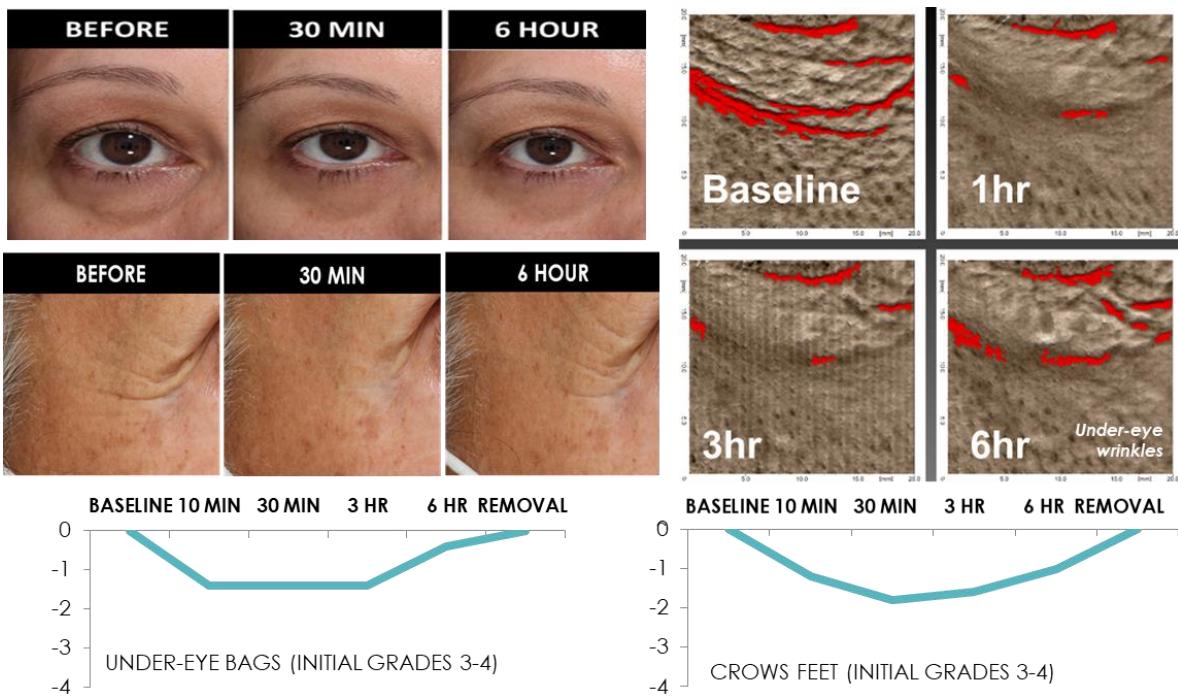


Figure 4 Improvement in undereye bags, wrinkles and crowsfeet after application of the film forming complex in an expert setting study for 6 h duration

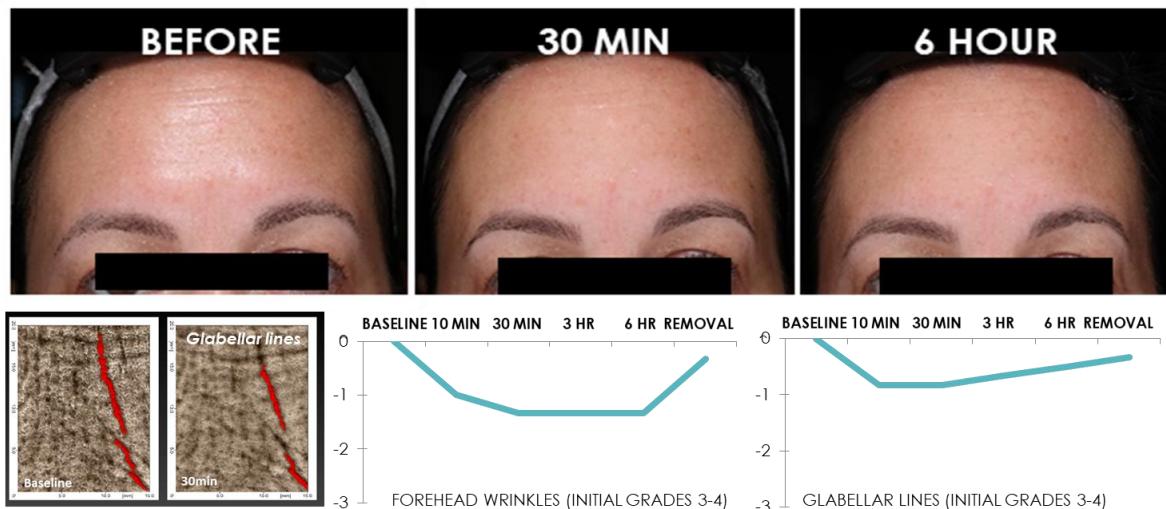


Figure 5 Improvement in forehead and glabellar wrinkles after application of the film forming complex in an expert setting study for 6 h duration

#### In Vivo Performance- Consumer Studies

The efficacy of the film-forming system in W/O emulsion was confirmed in 7 iterations of use-tests with about 500 consumers where they reported experiencing instant eyelift or botox-like effect

upon application of the formula on their undereye area and forehead, with slight tightening, compressing and blurring effect, with good comfort, lastingness, resistance to facial movements and easy removability.



*Figure 6 Consumer Selfies from a blind-use quantitative test*

In a large blind use quantitative study of the film-forming complex (n=116) against silicate technology (n=122), the consumers found the performance of the film-forming complex on undereye bags and wrinkles to be at par with the silicate technology but observed greater benefits from the film-former complex for comfort, lastingness, lack of flaws and compatibility with makeup. The proposal of dual application on forehead and undereye contour further heightened the interest and purchase intent of the consumers and the consumer had an enhanced sense of uniqueness of this technology. The current technologies (silicate and reactive silicone) are catered towards undereye usage and have severe incompatibility with make-up and hence the offering of multi-zonal application with ability to incorporate in their current skincare and/or makeup routine was appealing to the consumers.

*Table 1 Performance of the film-forming technology agaisnt silicate technology in a blind use quantitative test*

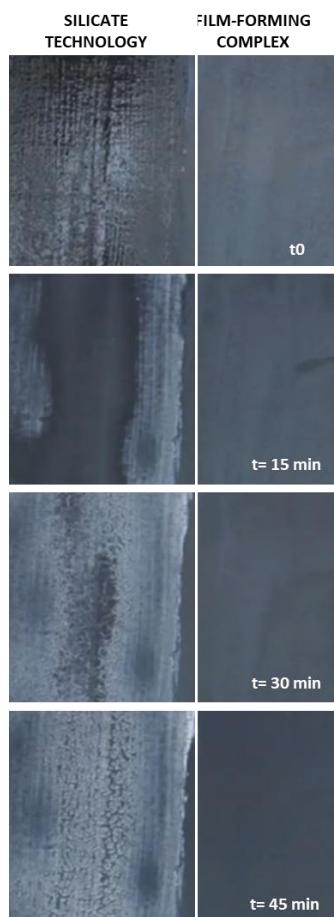
KEY PROJECT ATTRIBUTES	CONSUMER END BENEFITS	PERFORMANCE VS. SILICATE TECHNOLOGY
	Flaws ( <i>Whitening</i> )	+
	All-day lastingness	+
	Instant & dramatic results on undereye bags	=
	Improved appearance of eye contour	=
KEY CATEGORY DRIVERS	Comfort ( <i>during all-day wear</i> )	++
	Makeup compatibility	++

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**MUST-HAVE****Safety (*Irritation, redness*)****++**

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Another advantage of this film former technology is the lack of white cast that the consumers noted with silicate technology. The white-cast after silicate formula application is even more evident in those with deeper skin tones. This effect was replicated on a synthetic leather substrate by applying silicate formula and film forming complex and allowing the formulas to dry. A significant whitening and cracking was observed as the silicate film formed, unlike the matte clear film formed by the film-forming complex (Figure 7). Thus, film-forming technology provides a solution for transformative skin.



*Figure 7 Images of films of silicate formula and film forming complex on a synthetic leather substrate over 45 min*

## **Conclusion**

In conclusion, a film-forming complex with tunable physical and mechanical properties was engineered to cater to the consumers' demand for an at-home solution for instant and dramatic reduction of undereye bags, and eye/glabellar/forehead wrinkles. This technology proves to be a significant improvement over the currently available solutions for such applications for undereye area which include mainly silicate technology and a limited number of products with reactive silicone technology. The versatility of this technology offers the consumers the ability to easily incorporate it into their daily skincare or makeup routine and use it on multiple facial zones for a holistic rejuvenated appearance. This novel technology paves the way for future hybrid technologies at skincare-makeup interface, offering a new possibility for consumers to temporarily combat signs of fatigue and aging with easy application and removability.

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**Conflict of Interest Statement.** None.

## References

1. Yu B, Kang SY, Akthakul A, et al. An elastic second skin. *Nat Mater.* 2016;15(8):911-918. doi:10.1038/nmat4635
2. Jachowicz J, McMullen R, Prettypaul D. Alteration of skin mechanics by thin polymer films. *Skin Res Technol.* 2008;14(3):312-9. doi: 10.1111/j.1600-0846.2008.00296.x.
3. de Mul MNG, Uddin T, Yan X, Hubschmitt A, Klotz B , Chan WKM. Reducing facial wrinkle size and increasing skin firmness using skin care polymers, *J. Cosmet. Sci.*, 69 (2018), pp. 131-143
4. Maidhof R, Knapp E, Liebel F, Fair M, Rubinson EH. Technical approaches to select high-performance instant skin smoothing formulations: Correlation of in vitro and in vivo assessment methods. *Skin Res Technol.* 2019; 25: 606– 611.  
<https://doi.org/10.1111/srt.12691>
5. Masayuki Iida. Novel facial rejuvenation method: rapid removal of laugh lines using special cosmetic film . IFSCC 2020, Yokohama (online, October 21-30, 2020).

6. Method for reducing skin wrinkles, WO 2020067358A1 assigned to KAO (2020)
7. Portal J, Schultze X, Taupin S, Arnaud-Roux M, Bonnard J, Naudin G, Hely M, Bui H, Biderman N. Adhesion Aspect and Film-Forming Properties of Hydrocarbon Polymers-Based Lipsticks. *Surface Science and Adhesion in Cosmetics* 2021:451-48.  
<https://doi.org/10.1002/9781119654926.ch14>
8. Bernard A, Deng Y, Bui H, Daubersies L, Debeaud R, Farran A. Compositions and methods for improving the appearance of the skin, US patent 10,864,157 assigned to L'OREAL (2020)
9. Luengo G, Bui H and Portal J. Formation and Performance of Cosmetic Films in Cosmetics. *Handbook of Cosmetic Science and Technology*. Ed. Dreher F, Jungman E, Sakamoto K and Maibach H. CRC Press. 2022. 167-181.