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Preparation and evaluation of a cosmetic adhesive containing guar gum

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Abstract−Guar gum is an effective agent for use as a natural adhesive ingredient that can be used to replace the hazardous ingredients of spirit gum. This study describes the possibility of using guar gum as a renewable substituent for cosmetic adhesive. An adhesive base and cosmetic adhesive containing guar gum were prepared by a two-step process. The samples were tested for safety (cell toxicity test and patch test), stability (centrifugation, cycling and viscosity), and effectiveness (tensile strength). The results from the MTT assay show that the growth activity of human fibroblast skin cells was over 89% in all concentrations of cosmetic adhesive containing guar gum. In addition, no special skin reactions were reported in the patch test prepared adhesive containing guar gum. Moreover, the stability test demonstrated proper stability of all adhesive samples: the composition stability, heat stability, and viscosity of the adhesive samples maintained stable conditions. The efficacy test confirmed the superiority of the guar gum adhesive samples over spirit gum concerning the tensile strength. This study demonstrated that guar gum may be a viable replacement for synthetic rosins and also as a substituent in cosmetic.

Keywords: Guar Gum, Cosmetic Adhesive, Safety, Stability, Tensile Strength

INTRODUCTION

Spirit gum is the traditional skin adhesive used in the cosmetics industry. It has been manufactured since the 1890s and has been a standard tool in theatrical performances involving prosthetic makeup or affixed costuming. It is used mainly by actors to attach crepe hair, mustaches, beards, wigs, fake noses and bald caps to their head [1]. Spirit gum is made mostly of specially denatured alcohol, rosin, and colophony. However, the available data is insufficient to support the safety of denatured alcohol containing spirit gum [2]. Many studies have reported that rosin and colophony cause allergic contact dermatitis [3-5]. Despite side effects, spirit gum continues to be used as a cosmetic adhesive, because it has strong adhesive ability on skin. There has been renewed interest to substitute spirit gum with a hypoallergenic skin adhesive. Therefore, natural and hypoallergenic ingredients, replacing hazardous substances that cause many skin problems, are of interest. Among the hypoallergenic adhesive ingredients, cyanoacrylate (histoacryl glue; N-butyl-2-cyanoacrylate) is a well-established pressure-sensitive adhesive, and it has been used for endoscopic management of gastrointestinal variceal bleeding in the medical field [6,7]. Cyanoacrylate is less irritating to the skin, superior in environmental stability, and a high surface energy adhesive. Therefore, cyanoacrylate-based adhesives are now being used for a multitude of new applications. Despite these advantages, cyanoacrylate is not commonly used as a cosmetic ingredient.

Natural adhesive ingredients are human friendly, hypoallergenic materials [8] formulated to enhance the curing speed and adhesion performance. Guar gum is a natural adhesive material used in food, drugs and cosmetics and as a thickening, stabilizing, suspending, and binding agent in foods and beverages. Guar gum also acts as a binding and thickening agent in lotions and creams. It is stable at a pH range of 4.0-10.5 and is chemically stable. Little evidence supports any side-effects of guar on the skin, but gastrointestinal complaints, such as nausea, stomachache, or meteorism, have been reported [9]. It is highly probable that guar gum could be useful as natural adhesive in cosmetic adhesive products.

Skin adhesives have a combination of traits that are distinct from those general-purpose adhesive products. Most obviously, the safety of using a skin adhesive is important. It should have no systemic toxic effects after its absorption into broken or compromised skin, including no allergic sensitization, or irritation. Additionally, it must stick to a skin layer that may be shed, because human skin has a very rough surface with hair, folds, creases, sweat pores and oil glands, and wrinkles. The surface energy of skin is low, so adhesion is further compromised by contamination with water, oils, salts and loose debris. Consequently, a proper adhesion shows good adhesive ability on the skin and then can be removed easily. The replacement of rosin and colophony with natural and hypoallergenic adhesive ingredients in cosmetics may provide many safety benefits and good performance.

In this study, we prepared a cosmetic adhesive containing hypoallergenic adhesive ingredients (histoacryl glue; N-butyl-2-cyanoacrylate and guar gum) and cosmetic ingredients. We assessed human skin fibroblast activation (cell viability, MTT-assay) and performed a patch test to evaluate the safety of the adhesive, followed by centrifugation, cycling, and viscosity tests to evaluate the stability. Additionally, we tested tensile strength to evaluate the per-

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Table 1. Formulations of the adhesive base and cosmetic adhesive (unit: g)

Phase	Ingredient	Adhesive base	Cosmetic adhesive
A	Distill water	66.82	61.82
	Carboxymethyl cellulose	0.20	0.20
	Ethylene diamine tetraacetic acid disodium salt	0.03	0.03
	Glycerin	3.00	3.00
	Polyvinyl alcohol	14.80	14.80
B	Ethanol	9.00	9.00
	Polyvinylpyrrolidone	0.50	0.50
	Methylparaben	0.20	0.20
	Nonylphenyl ether	0.40	0.40
	Histoacryl	0.05	0.05
	Polybutene	5.00	5.00
C	Guar gum	-	5.00

formance of the adhesive.

MATERIALS AND METHODS

1. Materials and Preparation of Adhesive Containing Guar Gum

Guar gum (Sigma-Aldrich, St. Louis, MO, USA) as the natural adhesive ingredient, histoacryl as the skin adhesive ingredient, and distilled water, polyvinyl alcohol, ethanol, glycerin, polybutene, antioxidants (ethylene diamine tetraacetic acid disodium salt and methylparaben), carboxymethyl cellulose, polyvinylpyrrolidone, and nonylphenyl ether were used. The composition of the cosmetic adhesive prepared is shown in Table 1.

Phase A was prepared by dissolving 14.8 g polyvinyl alcohol, 3 g glycerin, 0.2 g carboxymethyl cellulose, and 0.03 g ethylene diamine tetraacetic acid disodium salt in 66.82 g distilled water (Fig. 1). Phase B consisted of 0.5 g polyvinylpyrrolidone, 0.2 g methylparaben, 0.4 g nonylphenyl ether, 0.05 g histoacryl, 5 g polybutene, and 9 g ethanol. Phase A was added to phase B to prepare the cosmetic adhesive base. Guar gum (5 g) was added to the cosmetic adhesive base and mixed using the Homomixer® (HM-U1.0, JPL Co, Seoul, Korea). Then, the base and base-containing guar gum were removed from heat and stirred at room temperature until the mixture congealed.

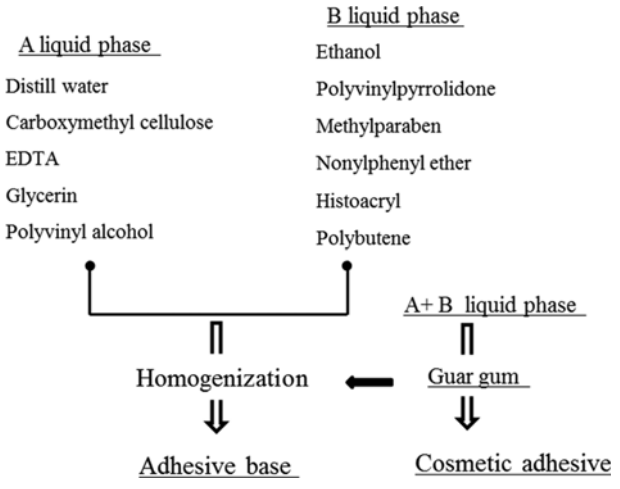

Fig. 1. Preparation process of adhesive base and cosmetic adhesive.

2. Cytotoxicity Assay

The cosmetic adhesive was tested in an *in vitro* cytotoxicity assay according to the ISO 10993 guidelines [11]. The cytotoxicity of the cosmetic adhesive was assayed *in vitro* using an MTT assay kit. CCD-986sk Human skin fibroblast cells (Korean Cell Line Bank 21947, Seoul, Korea) were cultured in Dulbecco's modified Eagle's medium supplemented with 10% fetal bovine serum and 1% antibiotics under 5% CO_2 at 37 °C. The fibroblasts were added to a 96-well plate at a density of 1×10^5 cells/well. The medium was replaced with fresh medium containing various concentrations of the cosmetic adhesive extract solution in 0.1 M PBS (pH 7.4) at 37 °C for 24 h. After incubation for 24 h, the cell cultures were mixed with the MTT assay reagent for 4 h, and the optical density was read on a multiwall microplate reader (BioTek Instruments Inc., Winooski, VT, USA) at 570 nm.

3. Patch Test

The cosmetic adhesive was evaluated by the human repeat insult patch test according to the methodology modified from Marzulli and Maibach [12]. Healthy, non-pregnant Korean women (age, 20-25 years) were screened, and 20 were enrolled. Skin on the upper back with an absence of scars, moles, freckles and any other skin anomalies was tested. The sample (25 μL, as is) was applied four times to the same site (induction site) over four consecutive weeks under an occlusive patch, followed by a challenge phase after a minimal one-week rest period. The patch test results were interpreted according to the International Contact Dermatitis Research Group (ICDRG) guidelines.

4. Determination of Stability (Centrifugation, Cycling and Viscosity)

Physical characteristics, including the composition stability, heat stability, pH and viscosity of the prepared cosmetic adhesives, were determined. Adhesive stability was measured using a centrifugation method (HA-1000-3 benchtop centrifuge; Hanil Science Medical, Daejeon, Korea). Test tubes were filled with 50 g samples in each and centrifuged at 1,238 g for 4 h.

The stability of the prepared cosmetic adhesive was determined

by hot-cool cycling from - 4 to 40 °C, with a 3 h storage time at each temperature using a multi-room temperature humidity incubator (DS-114; Dawon Science, Seoul, Korea). Seven cycles were performed, and the appearance of the cosmetic adhesive was observed at the end of each cycle. Stability was determined by the appearance, color and separation of the adhesive.

The viscosities of the samples were determined at 25 °C at 30 rpm using a Brookfield viscometer (DV-E Viscometer, Brookfield, MA, USA) with the LV spindle. The pH of the samples was monitored at room temperature with a pH-meter (sensitivity: ±0.255) (3110 WTW, Oberbayern, Germany) for 35 days.

5. Determination of Adhesive Ability

Pieces of cowhide were cut into sizes of 100×25×2 mm and 100×25×2 mm, respectively. The adhesive samples were applied between two pieces of cowhide and allowed to dry for 30 min. The tensile strengths of the adhesive samples were measured by an Instron universal testing machine (Model 5566, Instron Corp., Canton, MA, USA) at a crosshead speed of 2.0 mm/min. A force of 1,000 lb was applied to the load cell. Tensile strength was expressed in MPa and calculated by dividing the maximum load (N) by the initial cross-sectional area (m^2) of the specimen. This procedure was repeated five times for each type of sample.

6. Statistical Analysis

All statistical procedures were conducted by using SPSS ver. 21.0 software (SPSS Inc., Chicago, IL, USA). Data are presented as means and standard deviations. The results were subjected to one-way analysis of variance, and significant differences were determined by Duncan's multiple range test at $P<0.05$.

RESULTS AND DISCUSSION

1. Safety Test

We first assessed the safety of the adhesive samples (Fig. 2). The CCD-986sk human fibroblast skin cell line was treated with each sample at concentrations of 1,250, 2,500, 5,000, and 10,000 ppm and then subjected to MTT assay. The viability of human fibroblast skin cells decreased after exposure to increasing concentrations of spirit gum. A cytotoxic effect was observed at 5,000-10,000 ppm of spirit gum. Cell viability in response to the adhesive base and adhesive containing guar gum (cosmetic adhesive) was above 80% at all concentrations. Cosmetic adhesive showed higher cell viability at concentration of 10,000 ppm than that of the other samples.

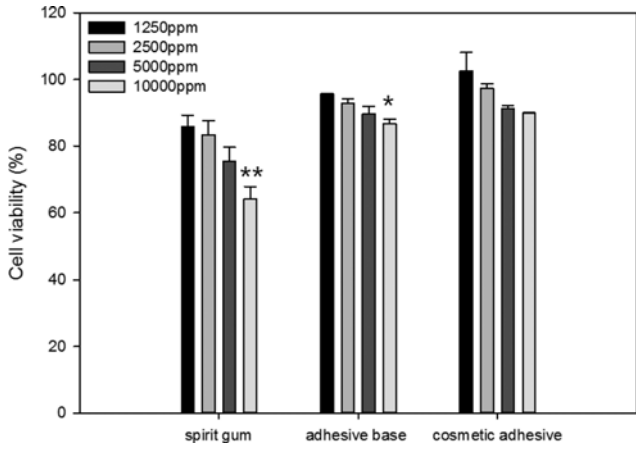

Fig. 2. Cell viability of spirit gum, adhesive base and cosmetic adhesive on the CCD-986sk cell (n=3, *P<0.05, **P<0.01).

Table 2. The results of patch tests in spirit gum, adhesive base and cosmetic adhesive

Sample	−	+/−	+	++	+++	Positive rate (%)
Spirit gum	19	0	1	0	0	1 (5%)
Adhesive base	20	0	0	0	0	0
Cosmetic adhesive	20	0	0	0	0	0

−: No reaction, −/+: Weak positive reaction (Erythema), +: Moderate positive reaction (Erythema, Induration), ++: Serve positive reaction (Erythema, Induration, Vesicles), +++: Serve positive reaction (Erythema, Induration, Bullae)

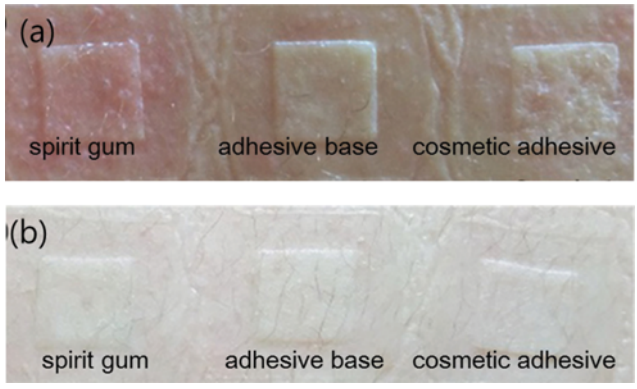

Fig. 3. Representative photographs of grade of skin reaction by human patch test. 1(+) grade from subject (red circle indicated the spirit gum) (a) and No reaction from subject (b).

The interpretation of the patch test results is very important for safety. The human patch test is not standardized, in contrast to *in vitro* tests or animal studies; however, various factors can affect the skin irritation response, such as personal characteristics and the season [13]. Accordingly, the patch test was conducted on 20 female volunteers and three adhesive samples were tested over four weeks (Table 2). No skin reaction was observed in response to the prepared adhesive base or adhesive containing guar gum, whereas one case showed a positive reaction (erythema) to spirit gum (Fig. 3). Colophony is a skin sensitizer and the third leading cause of occupational asthma [14]. Overall, modified products are generally stronger sensitizers. As a result, standard patch testing using unmodified rosins fails to detect patients allergic to chemicals in the modified rosins [15]. In a previous report, the rate of positive reactions to spirit gum was 3.3% [16]. Our safety test results agreed with those of previous studies and with expected results.

2. Stability Test

The adhesive samples were initially evaluated for physical stability using a centrifugation method. All samples maintained homogeneity. All samples were stored in a multi-room temperature humidity incubator at temperatures ranging from - 4 to 40 °C, and then stability was assessed by the appearance, color, and separation of the samples. All samples remained homogeneous under these

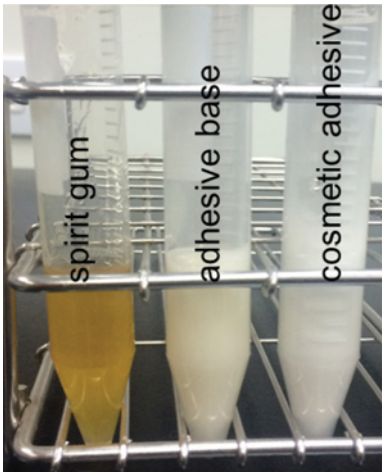

Fig. 4. Centrifugation test and cycling test of spirit gum, adhesive base and cosmetic adhesive.

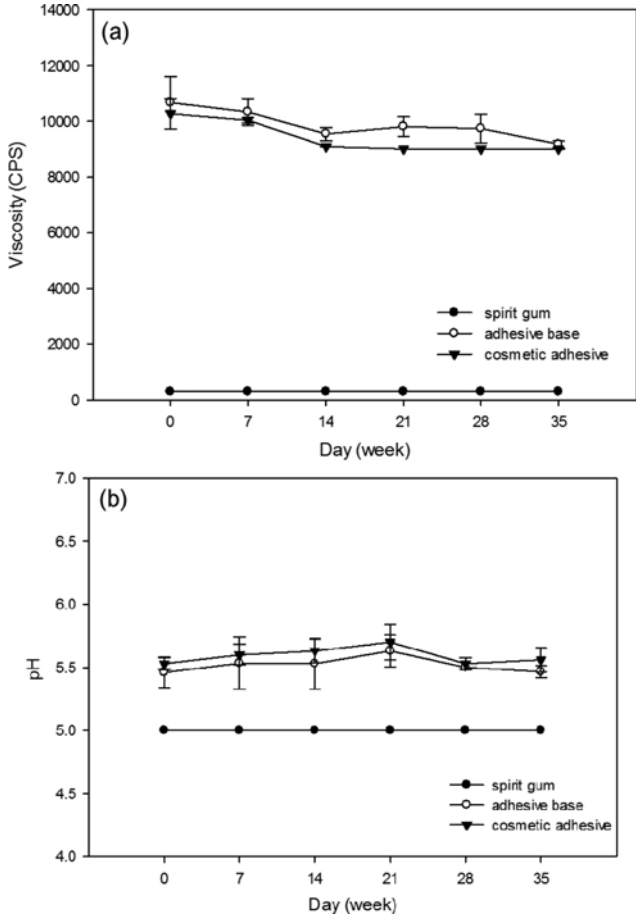

Fig. 5. Stability test of viscosity (a) and pH (b) for 35 days.

conditions and did not change as the temperature was increased (Fig. 4); therefore, the samples were considered to have proper stability.

The viscosity of the samples was measured at 25 °C for 35 days, as shown in Fig. 5(a). Spirit gum had a low viscosity of approximately 300 cps and assumed non-thickened liquid state. The prepared adhesives (the control and cosmetic adhesive) were thicker, with viscosities of 9,000-10,000 cps. The prepared adhesive containing guar gum remained a translucent fluid with decreased viscosity and gel-like behavior. The physical properties of the control and cosmetic adhesive were altered within 1,000 cps. However, the prepared adhesives had negligible effects on the physical properties. Cosmetic adhesive had the highest viscosity compared with the control and spirit gum. As mentioned above, as a thickening agent, guar gum entered the external phase of the adhesive base to increase its viscosity. A direct relationship exists between viscosity and stability [17], in that increased viscosity produces a more stable adhesive. The prepared adhesive containing guar gum was stable for use as a cosmetic adhesive.

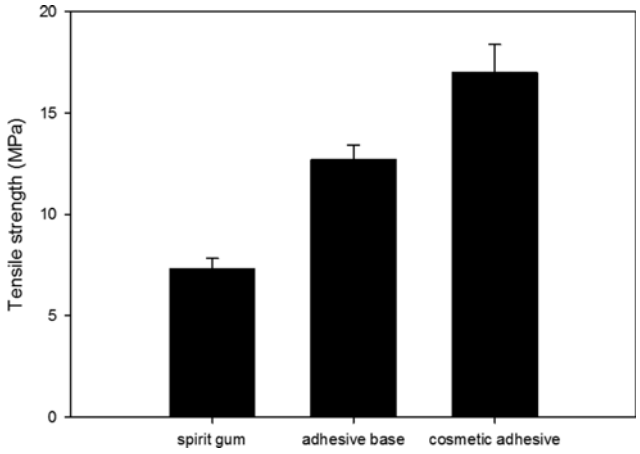

Fig. 6. Tensile strength of spirit gum, adhesive base and cosmetic adhesive.

Various skin pH values have been reported, all within the acidic range of pH 4.0 to 7.0. The mean pH of the skin surface is 5.0-6.0 [18]. No significant changes were seen in either formulation over the 35-day period, regardless of the storage duration, indicating that this formulation has suitable pH for topical application.

3. Efficacy Test

The tensile strength of the adhesive samples was measured to evaluate adhesive ability. A comparison of the adhesive strengths of the samples is shown in Fig. 6. The effectiveness of the prepared adhesives was superior to that of spirit gum. Cosmetic adhesive revealed the highest adhesive strength of 15.85 MPa. Adhesive strength increased as more guar gum was added, confirming that the natural adhesive ingredient enhances adhesion performance. Kim [19] reported the tensile strengths of various synthetic resins, such as Paraloid B-72 (0.18 N/mm^2), paraloid B-44 (0.82 N/mm^2), cemedine C (0.83 N/mm^2), caparol binder (1.29 N/mm^2), and Devcon 5 minute (3.29 N/mm^2). The strengths of the prepared adhesives in this study were within the range of those of general purpose adhesive. The skin adhesive exhibited proper adhesion and was removable after use. Therefore, the adhesive containing guar gum has strong potential for practical use.

CONCLUSIONS

A hypoallergenic cosmetic adhesive containing guar gum, his-

toacryl, polybutene, and other ingredients was prepared and tested for safety, stability, and effectiveness. The growth activity of human fibroblast skin cells was over 89% at all concentrations of the cosmetic adhesive containing guar gum, while the proliferation (%) was 49.93% in the 10,000 ppm concentration of spirit gum. Moreover, according to the patch test result, no positive skin reaction was detected in response to the prepared adhesive containing guar gum, whereas a 2.4% weak positive reaction to spirit gum was observed. The adhesive containing guar gum exhibited stable physical characteristics, including the pH, color, composition, temperature and humidity conditions, and viscosity. Furthermore, the efficacy test confirmed the superiority of the adhesive samples over spirit gum for peeling adhesion. These results indicate that the prepared cosmetic adhesive containing guar gum is safe and stable and exhibits superior adhesion. This study concludes that guar gum may be useful as a replacement for synthetic rosins in adhesives; thus, practical uses for adhesive containing guar gum in cosmetics are expected.

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REFERENCES

1. E. Sina, *Handbook of Adhesives and Surface Preparation: Technology, Applications and Manufacturing*, Elsevier, Amsterdam (2010).
2. P. M. Satturwar, P. M. Mandaogade, G. N. Darwhekar, S. V. Fulzele, S. B. Joshi and A. K. *Dorle, Durg Dev. Ind. Pharm.*, **29**, 669 (2003).
3. E. Gäfvert and G. Färm, *Contact Dermatitis*, **33**, 369 (1995).
4. C. R. J. Hood, R. S. Cornell and B. Greenfield, *J. Foot Ankle Surg.*, **55**, 209 (2016).
5. S. Doukaki, G. Pistone, M. Arico and M. R. Bongiorno, *Dermatitis*, **23**, 298 (2012).
6. Z. Czech and R. Kurzawa. *J. Appl. Polym. Sci.*, **106**, 2398 (2007).
7. F. Weilert and K. F. Binmoeller, *Gastroenterol. Clin. North Am.*, **43**, 807 (2014).
8. J. K. Oh, D. H. Lim, S. Y. Kim and H. J. Kim, *J. Adhes. Interf.*, **9**, 34 (2008).
9. W. Blaschek, S. Ebel, E. Hackenthal, U. Holzgrabe, K. Keller, J. Reichling and V Schulz, *Hagers Handbuch der Drogen und Arzneistoffe*, Springer Berlin Heidelberg, New York (2006).
10. I. Akyar, *Wide spectra of quality control*, Intech, Croatia (2011).
11. D. Y. Noh, S. H. Joe, H. U. Yang, D. G. Han, J. H. Kim and D. U. Kim, *Korean Chem. Eng. Res.*, **57**, 762 (2016).
12. F. N. Marzulli and H. I. Maibach, *Contact Dermatitis*, **2**, 1 (1976).
13. S. M. An, H. Ham, E. J. Choi, M. K. Shin, S. S. An and H. O. Kim, *Int. J. Cosmet. Sci.*, **36**, 62 (2014).
14. A. Mariano, I. Paredes, R. Nuti and A. Innocenti, *Med Lav.*, **84**, 459 (1993).
15. R. D. Quain, G. Militello and G. H. Crawford, *Dermatitis*, **18**, 96 (2007).
16. H. Y. Yeo, K. Park and J. H. Kim, *J. Soc. Cosmet. Sci.*, **17**, 381 (2011).
17. J. Viyoch, N. Pisutthanan, A. Faikeua, K. Nupangta, K. Wangtorpol and J. Ngokkuen, *Int. J. Cosmet. Sci.*, **28**, 125 (2006).
18. H. Lambers, S. Piessens, A. Bloem, H. Pronk and P. Finkel, *Int. J. Cosmet. Sci.*, **28**, 359 (2006).
19. E. K. Kim, D. W. Park and S. Y. Jang, *J. Adhes. Interf.*, **14**, 128 (2013).