

# Coating Properties of UV Curable Coatings on Natural Nails: Impact Evaluation of 5 Different Acrylate Monomers

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Compositions on natural nails mostly contain acrylic monomers, more often 2-hydroxyethyl methacrylate (HEMA). These systems have been considered to be a potent sensitizer in contact with the skin. The purpose of the study was to compare five acrylic monomers: HEMA (hydroxyethyl methacrylate), HPMA (hydroxypropyl methacrylate), IBOMA (isobornyl methacrylate), MEP (methacryloylethylphosphate) and TMPTMA (trimethylpropane trimethacrylate) to find the substitute to HEMA. In this study, we explored the rheological and polymerization properties of uncured systems containing five acrylate monomers. Mechanical and adhesion properties were evaluated of cured compositions. Optimal criteria of compositions and cured coatings were defined, and tests were performed to find the most appropriate composition. HEMA is the most widely used acrylic monomer in nail coatings due to its outstanding characteristics: high reactivity (temperature reached at polymerization  $T_{\max} = 95\text{ }^{\circ}\text{C}$ ), good adhesion (1.5 MPa on glass and aluminum), high solubility in acetone ( $m_s = 22\%$ ). The greatest potential to substitute HEMA has monomer HPMA, due to its good abrasion resistance ( $G = 66\text{ GU}$  after abrasion), high reactivity ( $T_{\max} = 88\text{ }^{\circ}\text{C}$ ), and adhesion (1.5 MPa on glass and on aluminum). The polymerization process of a composition containing MEP was the fastest and reached the highest maximum temperature during polymerization process ( $T_{\max} = 115\text{ }^{\circ}\text{C}$ ) and great adhesion (2.5 MPa). The best surface abrasion resistance ( $G = 76\text{ GU}$  after abrasion) was reached for coatings containing TMPTMA.

**Keywords:** coatings, acrylate monomers, adhesion, polymerization, nail coatings.

## 1. INTRODUCTION

During the last decades, UV curable systems and coatings have become very popular in the applications for human – in dentistry as dental fillings [1], in medicine and beauty care [2–5] as coatings on natural nails. From the environmental perspective, these systems are much better for the ecosystem compared to silver/ mercury dental fillings [6, 7] or solvent-based nail coatings [5, 8]. Still, there is a high risk of irritation, as these systems mostly contain acrylic monomers. These UV curable systems have been taken from the industry (paints, coatings), where the contact with the human body is not intended.

Acrylic monomers in applications, which are in contact with the skin, have been considered to be potent sensitizers, that can cause skin itching, peeling, redness, irritation and allergic contact dermatitis [9, 10].

The choice of raw materials is very important in the development of any sustainable product or process, as raw materials have a significant impact on the sustainability profile, product performance, process efficiency and cost. This is especially important for personal care products. There are several factors to consider when choosing an ingredient for gel polish formulations for natural nails. The main components controlling the physical and mechanical properties of gel polish compositions are oligomers and monomers, starting from the structure-property relationship to meet such critical requirements as a rapid cure, low curing temperature, elasticity, durability, surface hardness, ease of

removal and non-toxicity. In addition, to improve the environmental profile of the gel polish, such materials may need to be with a smaller irritation index or even better - derived from bio-renewable materials or other sustainable resources [11].

Due to the increased environmental awareness, compositions and technologies of industrial coatings have been modified in many ways. The first step of making coatings more environmentally friendly was done by changing solvent borne systems to water borne systems or systems without solvents. Huge progress is reached due to the exclusion of VOC. Also, there can be found first compositions containing water-based emulsions instead of solvent-based emulsions. [12].

There have been studies to make acrylic monomers friendlier to the environment and humans: studies, where acrylic monomers were combined with bio-based phenols and analytical tests of the cured coatings, confirmed that this alternative cross-linking system gives a high level of performance with a high cross-linking degree, high thermal stability, good adhesion properties and excellent impact resistance. Still, there is a field for the improvement of flexibility, that will improve the adhesion and mechanical properties of cured coatings [13].

The most novel research has investigated the modification of fluorinated acrylic monomers with polyacrylate latex particles to reduce the amount of fluorine-containing acrylic monomers. This would decrease the environmental pollution aspects and increase the economic benefits of the coating used. This research issues a new kind

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of environmentally friendly waterborne acrylic resin and presents a simple method for optimizing the performance of waterborne resins [14].

However, there are very few studies on gel polish products based on bio-renewable resources [15] or an aqueous polyurethane dispersion [16], or sustainable gel polish ingredients developed from acrylated epoxidized soybean oil [17].

There have been started studies to make the urethane acrylates more sustainable [11], but still, these ingredients are not yet available in the market.

There are some studies where more irritant acrylic monomers have been tested versus less irritant monomers [18-22]. Summarizing studies from the last 20 years, we found that the most often used ingredient, which can cause these skin reactions, is 2-hydroxyethyl methacrylate (HEMA). These studies showed that compared to the second most used acrylic monomer 2-hydroxypropyl methacrylate (HPMA), HEMA has almost 2 times higher risk of sensitization than HPMA [23]. Our latest research showed that HPMA containing systems are less irritant: cytotoxicity to keratinocyte cell culture was not observed, instead of compositions containing HEMA [23].

Still, HEMA has been used so widely due to its chemical and physico-mechanical properties. HEMA is the most reactive monomer that has been used due to its low viscosity, good reactivity, and good adhesion properties [24].

The goal of our study was to identify the substitute to HEMA and compare it with four other monomers mostly used in UV curable formulations for natural nails comparing rheological and physico-mechanical properties, and adhesion.

## 2. METHODOLOGY

### 2.1. Criteria

The characteristics and compositions of short-term UV curable coating applications on natural nails hadn't been widely reported yet [1, 24–29], but there are many studies about the irritation caused by these coatings [3, 4, 10, 18–22]. This is mainly due to the acrylic monomers used, which have been added to the system to dilute it. HEMA is known as the most often used monomer due to its ability to dissolve the system, due to its reactivity and good adhesion properties [2, 3, 9, 24]. Therefore, we tested the effect of 5 different monomers on the characteristics of gel polish compositions to find the replacement monomer to HEMA.

The formulation of appropriate UV curable nail coatings is critical to achieve the required characteristics (Table 1).

### 2.2. Materials

Five different monomers were tested in nail coating base composition prepared at Kinetics Nail Systems Ltd. As base materials were used urethane acrylate oligomer 60 % by weight (Urethane Dimethacrylate, Exothane 26, Esstech), photoinitiator 5 % (ethyl-2,4,6-Trimethylbenzoylphenylphosphine, Lucirin TPO<sub>L</sub>, Basf) and 35 % one monomer: HEMA (hydroxyethyl

methacrylate, Esstech), HPMA (hydroxypropyl methacrylate, Esstech), IBOMA (isobornyl methacrylate, Esstech), MEP (methacryloylethylphosphate, Miwon), TMPTMA (trimethylpropane trimethacrylate, Miwon). For pigmented coatings 2 % of TiO<sub>2</sub>, (Durlin) was added.

**Table 1.** Criteria defined for optimal UV curable nail coatings

Response	Goal limits
Viscosity, mPas	1000–5000
Polymerization temperature, °C	< 70
Cure depth unpigmented coating, μm	200–250
Cure depth pigmented coating, μm	170–250
Surface gloss, GU	> 70
Surface gloss after abrasion, GU	> 70
Adhesion, MPa	2–3
Loss of adhesion, min	> 4 (> 65 %)

### 2.3. Test methods

**Viscosity.** The viscosity of urethane acrylate compositions was measured with rheometer KNX5112 (Kinexus), spindle 25 at 25 °C. The average result from three measurements was taken. The standard deviation was 3–5 %.

**Coating layers preparation.** All ingredients were mixed by stirring at 1800 rpm for 4 min in Dispermill KK 250. The coatings were obtained by Film Applicator (BYK) in thickness of 200 μm. All coating samples were cured under UV/LED lamp (KP800LED, λ = 400–410 nm, YI Liang electron technology Co, China) for 60 s.

**Cure depth.** This parameter was tested for clear and pigmented systems to evaluate the monomer activity and coating reactivity. The coating was applied in 200 μm thickness on Hegman gauge grindometer, cured under UV/LED (KP800LED) lamp for 60 s. The oxygen inhibition layer, which was not polymerized, was removed with a cotton pad. The thickness of polymerized layer was determined.

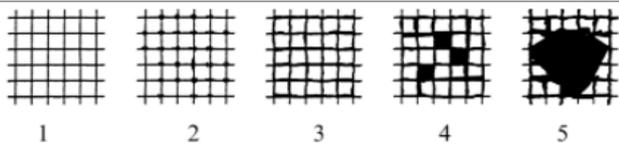
**Thermal analysis.** The exothermic effect of the polymerization process was studied. The coating material and inert reference were radiated under a UV lamp and the temperature difference between the sample and reference was plotted against the time. A K-type thermocouple was used to measure the temperature for reference (in the air) and in polymerized layer. Measurements were recorded by a data logger KTT-310 Meter Dual. The constant volume of the sample: 0.16 g ± 0.01 g, was weighted in the silicone with a working diameter of 10 mm. Both forms with thermocouples were placed under a UV lamp. Curves of *T* difference in time between 2 thermocouples were detected. The maximum temperature reached at polymerization was determined as *T*<sub>max</sub>.

**Surface abrasion.** The surface abrasion resistance of the samples was determined with a surface gloss meter and a surface scraping device developed at the laboratory of Kinetics Nail Systems. The coating was applied to the application sheet on 200 μm thickness, polymerized under a UV lamp for 60 s, and the unpolymerized layer was wiped with a cotton pad. The applied load was 3 kg acting on the surface with scraping cycle 680 scratches with sandpaper. The gloss of the coating before (*G*<sub>0</sub>) and after the abrasion test (*G*<sub>A</sub>) was determined with Novo Gloss Lite Gloss Meter

(Rhiopoint Instruments) on a black substrate at an angle of 60°.

**Pull-off adhesion.** The adhesion strength of the film was measured using a general paint and varnish test method (pull-off adhesion test from ISO 4624). Glass and aluminum panels were used as a substrate. The coating was applied on substrate as described previously, cured and left for 1 h to fully polymerize. After 1 h, the coating was slightly grounded with sandpaper and cleaned with 90 % acetone solution. The same was done with the contact surface of a metal dolly ( $D = 20$  mm). The contact surface of the dollies was covered with hot adhesive and applied to the top of the slightly sanded coating. The dollies were pressed against the glass for 30 s and left on the coating for 30 min to allow the glue to dry. After 30 min, a circle was cut around the dolly. The pull-off tester was screwed onto the top of the dolly and rotated clockwise until it pulled the dolly from the surface [25]. The test result was read at the moment when the pulling device lifted the dolly off the surface.

**Loss of adhesion.** The decomposition of the coating was evaluated using 95 % acetone. The uncured gel was applied on aluminum sheets with a thickness of 200  $\mu\text{m}$  (5 cm  $\times$  5 cm) and cured under a UV lamp for 60 s. The coatings were left for 72 h to allow the entire polymerization; the removability of the coating was then assessed. The coatings were wrapped in paper pads impregnated with acetone. The paper pads were coated with aluminum foil (100  $\mu\text{m}$  thick) and remained for 5 min. The adhesion of the coating to the substrate was measured with a cross-cut tester (BYK) using ISO 2409:2013 concept. After each 1 min period, the adhesion of the coating was assessed using adhesive tape (Scotch Crystal type 3M). The effective soak-off time was the time when 65 % of the coating has lost the adhesion to the substrate. The ISO 2409:2013 scale was used to evaluate the removability, Fig. 1 [26].



**Fig. 1.** Evaluation of loss of adhesion [26]

**Solubility of the coatings.** To evaluate the possible solubility of the coating, we extracted the polymerized films ( $m_0$ ) in a 95 % acetone solution. After 24 h extraction, we dried and weighted the films ( $m_1$ ) and calculated the extracted amount  $m_e$ :

$$\frac{m_0 - m_1}{m_1} \times 100\% = m_e \quad (1)$$

### 3. RESULTS AND DISCUSSION

**Viscosity.** Viscosity is the first criteria we test in new product development. Five monomer effect on base composition viscosity is given in Table 2. HEMA can dilute the acrylate oligomer base in the most efficient way. The second most effective monomer is HPMA and the less efficient is MEP. IBOMA and TMPTMA should be added in higher concentrations to achieve the viscosity which can be obtained with HEMA or HPMA.

**Table 2.** Monomer effect on composition viscosity depending on share rate

Monomer	Sear rate, $\text{s}^{-1}$		
	1	10	100
Viscosity, mPas			
Oligomer	7965	7902	7891
HEMA	1201	967	942
HPMA	1339	1179	1155
IBOMA	2271	2189	2127
TMPTMA	3662	3680	3611
MEP	7505	7513	7263

**Cure depth.** The next parameter tested was polymerization activity: polymerization rate and maximum temperature reached. The polymerization rate was determined by measuring the thickness of the polymerized coating for clear composition and pigmented composition with a Hegman gauge grindometer. Clear and pigmented coatings with 5 different monomers were applied on grindometer in 200  $\mu\text{m}$  thickness, cured under UV/LED lamp for 60 s. Non-polymerized oxygen inhibition layer was removed with a cotton pad. The thickness of the removed layer was determined. This parameter can characterize the monomer activity in the system. Data are given in Table 3. The higher is the value of cure depth, the more active is the polymerization in the thick layer.

**Table 3.** Monomer effect on clear and pigmented cured composition thickness

Unpigmented coating					
Content	HEMA	IBOMA	MEP	TMPTMA	HPMA
Cure depth, $\mu\text{m}$	200	200	200	200	200
Tacky layer, $\mu\text{m}$	< 5	< 5	< 5	< 5	< 5
Pigmented coating					
Cure depth, $\mu\text{m}$	200	200	180	175	200
Tacky layer, $\mu\text{m}$	< 5	< 5	< 10	< 10	< 5

The results show that monomer activity in the clear coating is identical, but in polymerization results of pigmented coatings we can see the difference. The more reactive compositions are those containing HEMA, HPMA and IBOMA.

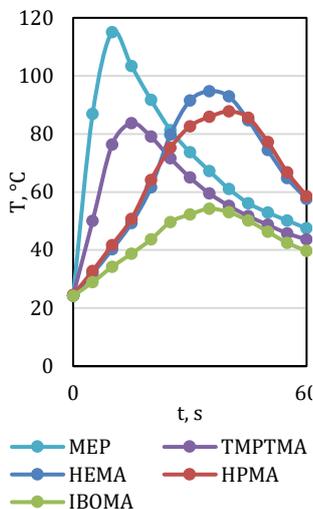
**Thermal analysis.** Thermal analysis was used to characterize these systems and the results are shown in Fig. 2. All systems except IBOMA exceed the limit of  $T_{\text{max}}$ . MEP and TMPTA reach the maximum polymerization temperature in the first 20 seconds of polymerization, but HEMA, HPMA and IBOMA reach the maximum temperature in the second part of polymerization time. The monomer concentration of MEP, TMPTA, HEMA and HPMA should be reduced to obtain a coating that meets the requirements.

**Surface abrasion.** Gloss and gloss retention of the coating in the wear time is another important parameter. We studied monomer effect on the gloss of the coating after application ( $G_0$ ) and after abrasion test ( $G_A$ ). Results are given in Figure 3. The coating gloss after polymerization is within the requirements for systems containing HEMA, HPMA, TMPTMA and IBOMA. After the abrasion test only TMPTMA containing coating meets the requirements.

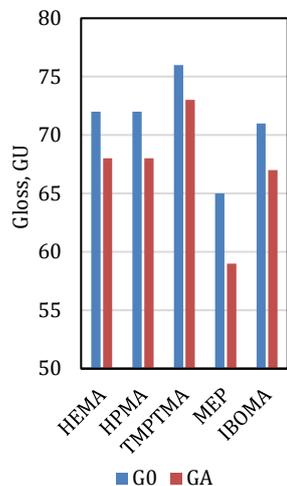
**Adhesion.** Monomer effect on coating adhesion was measured with pull-off method on the glass substrate and

aluminum substrate (which was detected previously as the best substrates for coating comparison). Obtained data are given in Fig. 4.

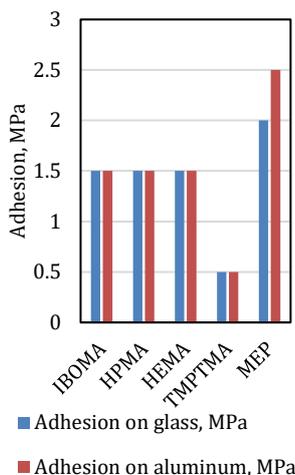
The best adhesion results have been achieved with systems containing MEP, and this was the coating where adhesion results on glass differ from adhesion results on aluminum.



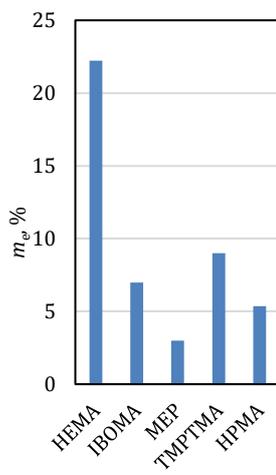
**Fig. 2.** Monomer effect on the composition polymerization temperature



**Fig. 3.** Monomer effect on coating gloss after curing ( $G_0$ ) and after abrasion test ( $G_A$ )



**Fig. 4.** Monomer effect on coating adhesion



**Fig. 5.** Effect of monomers on the amount of extracted coating

All other coatings have the same adhesion values on both substrates. HEMA, HPMA, IBOMA reached analogue adhesion values on both substrates (1.5 MPa), which do not fit in the required range. The worst adhesion was reached with monomer TMPTMA. These results correlate with coating removal time (Table 4). Only coatings containing MEP reach the defined criteria.

Solubility of the coating. Solubility of the coating is an important parameter to ensure gentle removal of the coating. In the base formulation there have been added some

additives to improve the solubility in acetone and loss of the adhesion.

**Table 4.** Monomer effect on loss of adhesion

Loss of adhesion time, min	IBOMA	HPMA	HEMA	TMPTMA	MEP
5	3	3	4	4	1
10	4	4	5	5	2
20	5	5	5	5	3
30	5	5	5	5	4

But still, there is also the effect of the added monomers. We compared the coating solubility in the acetone for 24 h and calculated the number of ingredients extracted after 24 h extraction in acetone. Results are given in Fig. 5. Compositions with HEMA showed the higher solubility compared to other systems. This fact explains the easy removal and short loss of adhesion time of the HEMA-containing systems.

Our research results support the fact, why HEMA is the most widely used monomer. HEMA has the most efficient ability to dissolve the composition, has high reactivity, average surface abrasion resistance, average adhesion, and high solubility in acetone to make the coating removal fast. Still, there are monomers like HPMA, IBOMA, TMPTMA and MEP which can be used to substitute HEMA in compositions with each other.

#### 4. CONCLUSIONS

The most widely used acrylate monomer in UV curing nail compositions is HEMA and our research results show that this monomer has the most efficient ability to dissolve the composition, HEMA has high reactivity (maximum  $T$  reached at polymerization  $T_{\max} = 115$  °C), average surface abrasion resistance ( $G = 68$  GU after abrasion test), average adhesion ( $A = 1.5$  MPa), and high solubility in acetone ( $m_e = 22$  %) to make the coating removal fast. The greatest potential to substitute HEMA has monomer HPMA, due to its high ability to dissolve the composition and get the required viscosity, due to its high reactivity ( $T_{\max} = 95$  °C), due to its surface abrasion resistance ( $G = 68$  GU after abrasion test) and adhesion ( $A = 1.5$  MPa). Still, there are possibilities to improve the characteristics of the coating adhesion by addition of MEP or to improve the surface abrasion by addition of TMPTMA. There is an opportunity to improve the coating properties by mixing the different monomers to make a composition that meets all criteria.

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