

# UV nanoimprint materials: Surface energies, residual layers, and imprint quality

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UV nanoimprint lithography is attracting more and more interest, because it has the potential of becoming a high-resolution, low-cost patterning technique. The availability of suitable UV curing materials is mandatory for successful imprinting. Within this work, a systematic investigation of commercially available photocuring materials was conducted to provide an overview of the properties of these materials. Their wetting behavior with respect to different substrate surfaces was characterized and their surface tensions were determined from their contact angles against two specifically selected solid surfaces: This method is presented here for the first time. The adhesion properties of the UV curing materials to different substrate surfaces and to the mold were investigated and necessary curing times were estimated. Additionally, the dependence of the residual layer thickness on the viscosity and the initial dispensed volume of UV curing materials was analyzed. It was found that the resist formulation of the UV curing materials strongly influences the surface tension as well as the adhesion to different substrate surfaces. Furthermore, the experiments verified that the thickness of the residual layer for UV curing materials increases with the square root of their viscosity which is predicted by theory. To demonstrate the suitability of the UV curing materials, first imprints with the prototype imprint tool, Nano Patterning Stepper 300 from Süss MicroTec, with pattern sizes down to 50 nm are shown. © 2007 American Vacuum Society. [DOI: 10.1116/1.2732742]

## I. INTRODUCTION

UV nanoimprint lithography (UV NIL) is attracting more and more interest as a technique to transfer nanosized patterns without using expensive optical exposure tools.<sup>1</sup> The resolution of this technique is only limited by the availability of patterns that can be resolved on a mold<sup>2</sup> and the availability of an appropriate UV curing material. This material has to fulfill several requirements such as low viscosity, low adhesion to the mold, good adhesion to the substrate, fast curing times, and high etch resistance to allow pattern transfer into the substrate. To achieve these properties, UV curing resist formulations are frequently acrylic based, but vinyl-ether-based or fluorinated-based materials have shown appropriate imprint properties as well.<sup>3–6</sup>

As already mentioned, the adhesion properties of UV curing materials play an important role. For the evaluation of adhesion properties, the influence of the interface mold/UV curing material and substrate/UV curing material has to be investigated. To achieve a good adhesion of the UV curing

material to the substrate, a very good wetting behavior and a good specific adhesion are necessary. The formation of a weak boundary layer has to be avoided. A weak boundary layer occurs, when chain molecules of the resist are mainly bonded to the substrate surface during the adsorption and, therefore, are no longer available for bindings to other molecules in the resist. This leads to reduced cohesion forces between the substrate and the resist. A second reason for the formation of a weak boundary layer is the formation of gas voids in the case of a bad wetting behavior of the UV curing material to the substrate. This results in defects within the cured resist layer. For these reasons, an appropriate UV curing material with a good wetting behavior to a specific substrate has to be chosen.

Within this work, different commercially available UV curing materials were investigated. First, the contact angles of the UV curing materials to clean silicon (Si) substrates, Si substrates primed with hexamethyldisilazane (HMDS), and Si substrates covered with the antireflective coating (ARC) DUV 112-6 from Brewer Science were measured. Secondly, the surface tension of the UV curing materials was determined by a method which is presented in this work for the first time. Thirdly, the adhesion properties of the UV cured materials to the three substrate surfaces and to the mold were inspected optically and compared.

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Beside the investigations on the adhesion properties, the influences of dispensed volume and viscosity on the residual layer thickness (RLT) were analyzed and compared to theoretical predictions.

Finally, first UV nanoimprints with the prototype imprint tool Nano Patterning Stepper (NPS) 300 from Süss MicroTec are presented to demonstrate the validity of the investigations.

## II. EXPERIMENTAL SETUP

### A. Imprint tool and experimental procedure

All imprints were performed using the imprint tool NPS 300 from Süss MicroTec. The exposure wavelength of the UV light emitting diode array is 375 nm and the power density can be ramped up to 120 mW/cm<sup>2</sup>. For the experiments, a power density of 30 mW/cm<sup>2</sup> was chosen to avoid an embrittlement of the UV curing materials. The UV curing materials were placed on the substrate surface manually using a pipette or automatically using a single drop dispenser just before starting the imprinting process.

200 nl of the UV curing resists were placed manually in the center of the imprint site and the following investigations were performed.

1. Estimation of the UV curing time for the UV curing materials.
2. Evaluation of the adhesion properties for the UV curing materials to the substrate surfaces and to the mold.
3. Verification of the theoretical predictions that the residual layer thickness increases with the square root of the viscosity of the UV curing material.

The single drop dispenser, where drops of the UV curing material are placed on substrate surfaces following a pattern predefined in the software of the imprint tool NPS 300, was used for (1) measuring the residual layer thickness for different volumes of UV curing material and (2) demonstration of first imprints with the NPS 300.

For all imprints, a fused silica mold with a size of 12.5 × 12.5 mm<sup>2</sup> and a surface roughness of less than 0.6 nm rms was used. The surface of the mold was modified with an antisticking layer [(tridecafluoro-1,1,2,2-tetrahydrooctyl)trichlorosilane] to provide a low surface tension and to allow a defect-free demolding from the cured material. During the imprinting process, the mold was pressed into the UV curing material following a predefined force profile. When the force reached its maximum level of 20 N, the material was cured by an UV exposure. After curing, the mold was separated from the material again.

For the determination of the residual layer thickness, the resist thickness of every imprint was measured at 24 points with a KLA Tencor P-2 long scan profilometer. The contact angles of the UV curing materials with the substrates were measured with a KRÜSS G-1 drop shape analyzer. From these measurements, the surface tensions of the UV curing materials were calculated.

TABLE I. Viscosities and experimentally determined UV curing times for the evaluated UV curing materials.

|                       |                                                                          | Viscosity $\mu$<br>(mPa s) | UV curing time<br>(s) at 30 mW/cm <sup>2</sup><br>and 375 nm |
|-----------------------|--------------------------------------------------------------------------|----------------------------|--------------------------------------------------------------|
| Dental UV<br>sealants | Fissurit (Voco)                                                          | >100                       | No curing                                                    |
|                       | Clinpro Sealant<br>(3M ESPE)                                             | >100                       | No curing                                                    |
|                       | Helioseal<br>(Ivoclar<br>Vivadent)                                       | 70–75                      | 100                                                          |
| UV glues              | NOA 61 (Norland<br>Optics)                                               | 300                        | 20                                                           |
|                       | Z-Resist (96.5%<br><i>t</i> -butyl acrylate<br>and 3.5%<br>Irgacure 369) | 12–14                      | <5                                                           |
| UV NIL<br>resists     | Inoflex RP+<br>(Inomat GmbH)                                             | 100                        | <5                                                           |
|                       | PAK 01 (Toyo<br>Gosei)                                                   | 50                         | <5                                                           |
|                       | NIF 1 (Asahi<br>Glass Company)                                           | 14.5                       | 10                                                           |
|                       | NIF 2 (Asahi<br>Glass Company)                                           | 24.5                       | 10                                                           |

### B. UV curing materials

Nine commercially available UV curing materials were investigated ranging from dental UV sealants and UV glues to dedicated UV NIL resists. The UV curing materials distributed by Molecular Imprints such as MonoMat™ were not available at the time of this work. The evaluated resists and their main properties are summarized in Table I. The UV curing times for an UV intensity of 30 mW/cm<sup>2</sup> were determined experimentally. The UV curing times range from less than 5 s for the Z-Resist, Inoflex RP+, and PAK 01 to no curing after 2 min for Fissurit and Clinpro Sealant. The UV resists, Fissurit and Clinpro Sealant, which did not show any curing after 2 min, were not considered for further investigations.

The dental UV sealants Fissurit, Clinpro Sealant, and Helioseal were obtained from VOCO, 3M ESPE, and Ivoclar Vivadent, respectively. The UV glue NOA 61 was provided by Norland Optics and Z-resist was a homemade mixture from *t*-butyl acrylate (96.5%) and the photoinitiator Irgacure 369 (3.5%) from CIBA. UV NIL resists Inoflex RP+ and PAK 01 were obtained by Inomat GmbH and Toyo Gosei, respectively. NIF 1 and NIF 2 are products of Asahi Glass Company.

### C. Determination of the surface tension of UV curing materials by contact angle measurements

To characterize the wetting behavior of solid surfaces, the measurement of the contact angle and the calculation of the surface tension are widely used. The surface tension ( $\sigma$ ) is

TABLE II. Surface tensions of Teflon and fused silica. The two solid materials were used to measure the surface tensions of the UV curing materials.

|              | $\sigma_{\text{polar}}$ (mN/m) | $\sigma_{\text{dispersive}}$ (mN/m) | $\sigma_{\text{total}}$ (mN/m) |
|--------------|--------------------------------|-------------------------------------|--------------------------------|
| Teflon       | <0.05                          | 27.5                                | 27.5                           |
| Fused silica | 94.7                           | <0.05                               | 94.7                           |

separated in a polar ( $\sigma^P$ ) and a dispersive part ( $\sigma^D$ ), where the polar part describes the hydrophilic character and the dispersive part the hydrophobic character of a surface,

$$\sigma = \sigma^P + \sigma^D. \quad (1)$$

For the calculation of the surface tension for solids, droplets of different liquids with known surface tensions are brought into contact with the solid surface under test. The interaction between the molecules in the liquid, the vapor phase, and the molecules on the substrate surface results in a droplet with an individual shape. The contact angle can be measured at the triple point liquid/solid/vapor phase. If there is a strong interaction between liquid and solid, a small contact angle occurs. A small contact angle means good wetting behavior and this is a prerequisite for a good adhesion.<sup>7</sup> Beside the chemical interaction, also the mechanical interaction influences the contact angle requiring a surface with a defined and homogeneous roughness for reproducible measurements.

The measurement of the surface tension for liquids is quite different. A tensiometer is typically used to determine the dispersive and polar parts of the surface tension by the ring or plate method.<sup>8,9</sup> In the following, a method will be presented for the first time to calculate the surface tension of liquids by measuring the contact angle of a liquid against two different solid surfaces. The advantage of this new method is that the surface tension of both, solids and liquids, can be measured with one conventional contact angle measurement tool. For this method, it is of great importance that the two solid surfaces are of high mechanical quality, e.g., that they have an optically polished surface to minimize influences of surface roughness. The calculation of the surface tension for the UV curing materials is based on the equation introduced by Owens and Wendt,<sup>10</sup>

$$\sigma_{sl} = \sigma_s + \sigma_l - 2\sqrt{\sigma_l^D \sigma_s^D} - 2\sqrt{\sigma_l^P \sigma_s^P}. \quad (2)$$

Here,  $\sigma_{sl}$  is the surface tension between the solid and the liquid material, and  $\sigma_s$  and  $\sigma_l$  are the surface tensions for the solid and the liquid material, respectively. The superscripted indices  $D$  and  $P$  describe the dispersive and polar parts of the surface tension. Combination of Eq. (2) with Young's Eq. (3),<sup>11</sup>

$$\sigma_s = \sigma_{sl} + \sigma_l \cos \Theta, \quad (3)$$

where  $\Theta$  describes the contact angle at the triple point solid/liquid/vapor phase, results in the following:

$$\sigma_l(1 + \cos \Theta) = 2\sqrt{\sigma_l^D \sigma_s^D} + 2\sqrt{\sigma_l^P \sigma_s^P}. \quad (4)$$

If both the polar part of the surface tension of a first solid material and the dispersive part of the surface tension of a second solid material are neglectable, Eq. (4) can be reduced to the following:

$$\begin{aligned} \frac{1 + \cos \Theta_1}{2\sqrt{\sigma_{s1}^D}} &= \frac{\sqrt{\sigma_l^D}}{\sigma_l^P + \sigma_l^D} = \nu_1 \\ \Rightarrow \sigma_l^P &= \frac{\sqrt{\sigma_l^D}}{\nu_1} - \sigma_l^D \end{aligned} \quad (5)$$

and

$$\frac{1 + \cos \Theta_2}{2\sqrt{\sigma_{s2}^D}} = \frac{\sqrt{\sigma_l^P}}{\sigma_l^P + \sigma_l^D} = \nu_2. \quad (6)$$

Indices 1 and 2 describe the first and the second solid material, respectively.  $\nu_1$  and  $\nu_2$  are introduced to reduce the complexity of the following equations.

For calculating the dispersive part of the surface tension for the UV curing materials, Eqs. (5) and (6) have to be combined.

$$\sigma_l^D = \nu_1^{-2} \left[ \left( \frac{\nu_2}{\nu_1} \right)^2 + 1 \right]^{-2}. \quad (7)$$

The polar part of the surface tension of the liquid can be calculated from Eqs. (7) and (4).

$$\sigma_l^P = \nu_1^{-2} \left\{ \left[ \left( \frac{\nu_2}{\nu_1} \right)^2 + 1 \right]^{-1} - \left[ \left( \frac{\nu_2}{\nu_1} \right)^2 + 1 \right]^{-2} \right\}. \quad (8)$$

As already mentioned, one solid surface has to be a polar surface and the other a dispersive surface, respectively. The dispersive surface used was optically polished Teflon with a dispersive surface tension of 27.5 mN/m and a polar surface tension below 0.05 mN/m. The polar surface used was fused silica (Lithosil L1 from Schott Lithotec) with a polar surface tension of 94.7 mN/m and a dispersive surface tension below 0.05 mN/m. The corresponding values are summarized in Table II.

### III. RESULTS AND DISCUSSION

#### A. Adhesion properties of UV curing materials

To achieve a good adhesion, a good wetting behavior of UV curing materials to solid surfaces is necessary but not sufficient. A good wetting behavior is obtained when the contact angle is small. In Table III, the contact angles of the evaluated UV curing materials for Si substrate, Si substrate primed with HMDS, Si substrate covered with an ARC, and the mold are summarized. In every case, the contact angles of the UV curing materials to the substrates are smaller than to the mold indicating a better adhesion of the UV curing materials to the substrates than to the mold.

In Table IV, the adhesion properties of the UV curing materials after the separation of the mold from the cured resist are summarized. After the imprint, the whole imprinting area on the substrate was inspected with an optical microscope and categorized. The entire resist stuck on the sub-

TABLE III. Contact angles of the evaluated UV curing materials for the three substrates and the mold (n.i.: not investigated).

|             | Contact angle (°) |         |        |      |
|-------------|-------------------|---------|--------|------|
|             | Si                | Si/HMDS | Si/ARC | Mold |
| NOA 61      | 23.7              | 41.8    | n.i.   | 68.3 |
| Helioseal   | 15                | 26.7    | n.i.   | 66.2 |
| Inoflex RP+ | 14.9              | 31.9    | 8      | 67.9 |
| PAK 01      | 9.4               | 20.3    | 8.7    | 66.5 |
| NIF 2       | 16.3              | 12.3    | 6.1    | 33.7 |
| NIF 1       | 15.8              | 11.4    | 5.9    | 34.2 |
| Z-Resist    | 6.3               | 25.8    | 10.6   | 65.4 |

strate after the imprint, when the combination substrate/resist is described in Table IV with “++.” If the resist stuck on the substrate to about 80%, 60%, 40%, and less than 40%, it is described with “+,” “0,” “–,” and “––,” respectively. The resists Helioseal, PAK 01, and Z-Resist showed very good adhesion properties to the clean Si substrate surface. Priming of the Si substrate with HMDS resulted in a reduced adhesion for the resists Inoflex RP+ and Z-Resist. UV curing materials Inoflex RP+, PAK 01, NIF 1, and NIF 2 showed very good adhesion properties to the Si substrate covered with an ARC.

Between the Si substrate and the resists NIF 1 and NIF 2, no adhesion was observed. This arises from the chemical formulation of these resists. They consist of a content of approximately 65% of fluorinated compounds. These fluorinated compounds decrease the specific adhesion to Si substrates. The resist Inoflex RP+ contains about 5% of fluorinated compounds. This amount is already sufficient to decrease the adhesion to Si substrates significantly.

In Table V, the surface tensions of the evaluated UV curing materials determined with the new method described in Sec. II C are shown. The resists Helioseal, Inoflex RP+, PAK 01, and Z-Resist show nearly the same values for their surface tensions. All these resists have an acrylic-based resist

TABLE IV. Summary of the adhesion properties of UV curing materials after the separation of the mold from the cured resist. If the entire resist stuck on the substrate after the separation, it is displayed with “++.” If the resist stuck on the substrate to about 80%, 60%, 40%, and less than 40%, it is described with “+,” “0,” “–,” and “––,” respectively (n.i.: not investigated).

|             | Si | Mold | Si/HMDS | Mold | Si/ARC | Mold |
|-------------|----|------|---------|------|--------|------|
| Helioseal   | ++ | --   | n.i.    | n.i. | n.i.   | n.i. |
| Inoflex RP+ | +  | –    | 0       | 0    | ++     | --   |
| PAK 01      | ++ | --   | ++      | --   | ++     | --   |
| NIF 2       | -- | ++   | 0       | 0    | ++     | --   |
| NIF 1       | -- | ++   | 0       | 0    | ++     | --   |
| Z-Resist    | ++ | --   | –       | +    | --     | ++   |

TABLE V. Surface tension of the evaluated UV curing materials.

|             | $\sigma_{\text{polar}}$ (mN/m) | $\sigma_{\text{dispersive}}$ (mN/m) | $\sigma_{\text{total}}$ (mN/m) |
|-------------|--------------------------------|-------------------------------------|--------------------------------|
| Helioseal   | 16.1                           | 24                                  | 40.1                           |
| Inoflex RP+ | 16.6                           | 24.2                                | 40.8                           |
| PAK 01      | 15.6                           | 23.5                                | 39.1                           |
| NIF 2       | 4.8                            | 17.4                                | 22.2                           |
| NIF 1       | 4.8                            | 17.4                                | 22.2                           |
| Z-Resist    | 14.8                           | 23.3                                | 38.1                           |

formulation. The resists NIF 1 and NIF 2 show a reduced surface tension resulting from the fluorinated compound resist formulation.

## B. Residual layer thickness

The formation of a residual layer during the imprinting process is unavoidable. To minimize the thickness of the residual layer is of great importance because the layer has to be removed by a plasma etching process prior to the pattern transfer into a substrate. The plasma etching process results in a loss of dimensional accuracy and has to be minimized. The time to reach the final residual layer thickness can be calculated by the following:<sup>12</sup>

$$t_f = \frac{\mu s^2}{2p} \left( \frac{1}{h_f^2} - \frac{1}{h_0^2} \right). \quad (9)$$

Here,  $t_f$  is the imprinting time, and  $h_0$  and  $h_f$  are the initial and the final resist thickness, respectively,  $p$  is the imprinting pressure,  $s$  is the distance the resist flows, and  $\mu$  is the viscosity of the resist. Within this work the influence of the parameter viscosity and initial resist volume on the residual layer thickness was investigated. In case of the initial thickness being much higher than the final thickness, Eq. (9) can be reduced to the following:

$$h_f = \sqrt{\frac{s^2}{2pt_f}} \sqrt{\mu} \\ \Rightarrow h_f \sim \sqrt{\mu}. \quad (10)$$

To assure that the initial thickness is much larger than the final thickness, 200 nl of resist were dispensed using a pipette on a Si substrate just before the imprinting processes started. With this method, the initial thickness was about a factor of 100 higher than the final thickness. The mold used did not have patterns on it to assure reproducible imprint conditions.

In Fig. 1, the residual layer thickness is shown for the UV curing materials Z-Resist, PAK-01, Helioseal, Inoflex RP+, and NOA 61. The residual layer thickness increases with the square root of resist viscosity as predicted in Eq. (10). With higher viscosities, the homogeneity of the residual layer decreases because of the reduced flow of the highly viscous resist.

In order to determine the influence of the initial dispensed volume on the residual layer thickness, in principle, resists

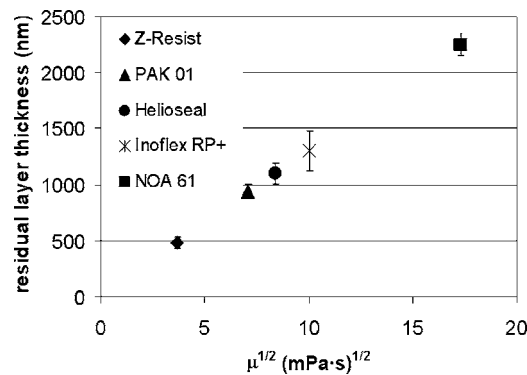


FIG. 1. Residual layer thickness for the UV curing materials.

with the lowest viscosities should be used as well as a dispense system to generate uniform drops with small volumes that cannot be realized manually with a pipette. As the dispense system currently used is not capable to process liquids with very low viscosities, the resists NIF 1 and Z-Resist, best suited for this kind of experiment, could not be used. Instead, the resist PAK 01 was used. The resist was placed on a Si substrate using the single drop dispenser following a pre-defined pattern that covers the whole imprinting area just before the imprinting cycle starts. The amount of resist was increased by dispensing the pattern one, two, three, and five times, respectively. The resulting residual layer thickness after the imprinting cycle was measured with a KLA Tencor P-2 long scan profilometer.

In Fig. 2, the resulting residual layer thickness for different volumes of PAK 01 is shown. The resulting residual layer thickness increases with a factor of about 2.5 when dispensing the pattern more than one time. The minimum residual layer thickness achieved for the resist PAK 01 was 400 nm which is not sufficient for a feasible pattern transfer of nano-sized patterns into a substrate. We estimate that the residual layer thickness can be reduced to a feasible value of less than 100 nm by using a patterned mold in combination with a low viscosity resist such as NIF 1.

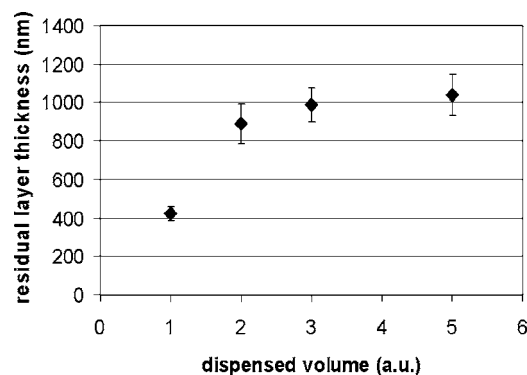


FIG. 2. Residual layer thickness for different dispensed resist volumes. The used resist was PAK 01.

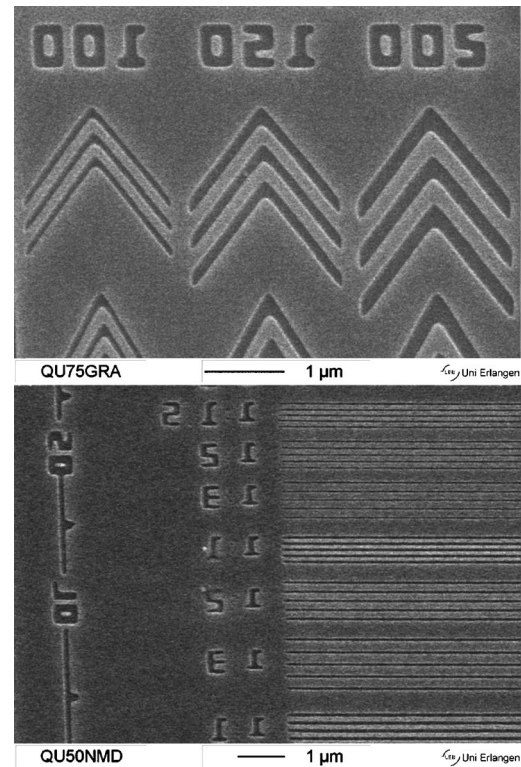


FIG. 3. SEM images of first UV nanoimprints with the imprint tool NPS 300. On the upper image, pattern sizes from 100 to 200 nm, and on the lower image, pattern sizes from 50 to 70 nm are transferred into PAK 01.

### C. First imprints

To demonstrate the capability of the imprinting tool NPS 300 and the validity of the investigations, first imprints into the UV curing material PAK 01 were performed. The resist pattern mentioned above was dispensed one time on a Si substrate and the curing time was 5 s at 30 mW/cm<sup>2</sup>. After the separation of the mold from the cured resist, no resist stuck on the mold and no defects were observed. In Fig. 3, scanning electron microscopy (SEM) images demonstrate the successful transfer of different patterns into the UV NIL resist PAK 01. The minimum resolved pattern size shown on the lower image of Fig. 3 was 50 nm and was limited by the size of the patterns on the mold. The successful imprints demonstrate the capability of the imprint tool NPS 300.

## IV. CONCLUSION

Within this work, different commercially available UV curing materials were investigated. Their contact angles to different substrate surfaces were measured and their surface tension was determined with a method presented here for the first time. The adhesion properties of the UV curing materials to different substrate surfaces and to the mold were discussed and their curing times were determined. Additionally, the dependence of the residual layer thickness on the viscosity and the initial dispensed volume of UV curing materials was demonstrated. It was found that UV curing materials that consist in parts of fluorinated compounds do not show appropriate adhesion properties to Si substrates or to Si sub-

strates primed with HMDS. The use of an ARC on the Si substrates results in an appropriate specific adhesion and, therefore, in acceptable adhesion properties. Except of the Z-Resist, all investigated UV curing materials show a very good adhesion to the Si substrate covered with an ARC. For the Z-Resist, a clean Si surface shows the best adhesion property. The dependence of the residual layer thickness on the viscosity and the initial dispensed volume was demonstrated. Theoretical predictions that the residual layer thickness increases with the square root of the viscosity of the resist could be approved. Finally, first UV nanoimprints with the tool NPS 300 could be successfully demonstrated.

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