

Research Article

Continuous Fabrication of Wide-Tip Microstructures for Bio-Inspired Dry Adhesives via Tip Inking Process

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Received 26 August 2018; Accepted 4 December 2018; Published 2 January 2019

Guest Editor: Hiroshi Umakoshi

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In this paper, we report a new method for continuous fabrication of dry adhesives composed of microstructures with mushroom-shaped ends. Conventional mushroom microstructure fabrication is performed with a simple molding technique using a reversed phase master. In a typical fabrication process, thin- and wide-tip portions may be ripped during demolding, making it difficult to use in a continuous process. It is also difficult to apply the mushroom structure master to a continuous process system in roll form. Here, a continuous fabrication process was developed by applying the method of fabricating a wide tip using a tip inking method after forming a micropillar. Through the continuous process, the dry adhesive was successfully fabricated and the durability was measured with a reasonable pull-off strength (13 N/cm^2). In addition to the reasonable adhesion, high durability is guaranteed, and fabricated dry adhesives are expected to be used in various fields.

1. Introduction

Mushroom-shaped microstructure-based dry adhesives are expected to be applied in various fields, thanks to their unique properties, such as strong adhesive force, repeatability, durability, reversibility, and self-cleaning [1–9]. The application is evident not only in daily necessities but also in industries as a very strong adhesive on a smooth surface. The principle and properties of adhesion of dry adhesives are widely known through the body structure of various living organisms [10–12]. However, in order for dry adhesives to be widely used, the efficiency of the production process needs to be increased. Until now, dry adhesives have been made by fabricating a silicon master using a complex surface micromachining process and then replicating thermosetting or UV-curable resins [1–13]. In most dry adhesives, because of the larger spatula or mushroom shape, it was very difficult and costly to fabricate a silicon master [4, 14]. Even if the master was produced, it was difficult to effectively reproduce through soft lithography or nanoimprint. Because of the wide-tip structure, the tip was torn during the demolding process and the microstructure was often broken. The limitations of the production method

suggest that continuous production techniques that can increase productivity are considered to be more difficult. Recently, processes for realizing such continuous production have been developed. First, the existing roll-to-roll imprint system was applied to produce UV-curable resin with a mushroom structure to produce dry adhesives [7]. Most ultraviolet-curable resins have a very high elastic modulus after curing, making it difficult to ensure adhesion. In order to solve this problem, a UV-curable resin with a relatively low elastic modulus was used. As a result, a dry adhesive having a reasonable adhesive strength could be continuously produced. However, this method is very difficult to produce a mold with a negative mushroom shape in the form of a film, and further development is required for high-yield production. In order to overcome these material limitations, a continuous production process using thermosetting polymers has been developed [5]. Polydimethylsiloxane (PDMS), which is mainly used for dry adhesives, has a very good conformal contact property and has excellent adhesion performance when it is made with dry adhesives. This is because it has a low elastic modulus. By fabricating the structure, higher contact performance can be realized because low effective elastic modulus can be

achieved with these structures. However, since this material generally takes about 1 hour to cure, continuous production is difficult to achieve. Lee et al. have developed a technology that can produce a continuous roll-to-roll type by thinning the sample and increasing the curing temperature to shorten the time [5]. In order to produce a dry adhesive having a wide tip for high-performance adhesives, the resin must be completely filled with a mold having a negative tip structure. However, since the PDMS has a high viscosity, ~ 2000 cPs, it is difficult to continuously fill the resin. Also, both these methods can be used as general promising techniques to fabricate dry adhesives. The fabricated master mold can be used to easily produce a dry adhesive with a simple molding technique after a simple release layer treatment; however, it is hard to apply it in a roll-to-roll-based continuous production process due to some failures during demolding (e.g., tip tearing depending on the polymer used) [15]. Recently, a continuous production technique using a two-step UV-assisted molding process has been announced [16]. A simple columnar microstructure is produced by the roll-to-roll imprint technique, but the tip portion is partially cured by appropriately controlling the curing time and UV intensity. Subsequently, a mushroom-shaped microstructure was developed by forming a wide tip on the column through the subsequent pressing and planarizing process. This method eliminates the need for a complex master fabrication process and avoids any failure that can occur with demolding. Since the thickness of the uncured resin that can be used for tip widening is only a few microns through the partial curing process, there is a limit to make a wide tip. In this respect, dry adhesives made through a partial curing process are limited in application because wide tips are essential to ensure strong adhesion of dry adhesives.

In this paper, we introduce a simple micropillar structure and a method of forming additional parts by inking the tips. This fabrication method can also be applied to the roll-to-roll-based apparatus without any severe changes; thus, it is applied to the prototype roll-to-roll production equipment to test the continuous fabrication possibility. Moreover, the pull-off strength of the bio-inspired dry adhesive was measured. The measured adhesion strength of approximately 13 N/cm^2 is slightly lower than that of dry adhesives made by typical one-step molding with PDMS; nevertheless, this value is applicable for application. It also has an advantage when compared with the continuous process using partial curing mentioned above. The adhesion strength of fabricated dry adhesives is lower when PDMS material is used but not significantly lower when the same soft polyurethane acrylate (PUA) is used. In addition, higher adhesion can be achieved if the tip size can be widened in future optimization processes.

2. Materials and Methods

2.1. Polyurethane Acrylate (PUA). The PUA consisted of a functionalized prepolymer with acrylate groups for cross-linking, a monomeric modulator, a photoinitiator, and a

radiation curable releasing agent (TEGO Chemical Service) for surface activity. The liquid mixture was drop-dispensed onto a silicon master with slanted pillars, which were prepared by an angled etching technique. A PET film with 100 μm thickness was gently placed on the liquid mixture, followed by UV exposure for 40 s (wavelength = 250–400 nm, dose = 100 mJ/cm^2). After UV curing, the mold was peeled off from the master, thereby leaving behind the PUA micromold. Normal PUA for mold and soft PUA for tip have 200 MPa and 20 MPa of elastic modulus, respectively.

2.2. Tip Widening. A feeding roller was used to induce the continuous supply of resin to soft PUA at the end of the fabricated micropillar. A doctor-blade-type resin dispenser was used to coat the soft PUA on the feeding roller and the pressure was applied for coating (~ 10 kPa). We also operated the roll device at a relatively slow speed to achieve sufficient inking (10 cm/min). After inking, the liquid resin was spread across the pressure roller and the wide tip was completely formed with additional UV exposure.

2.3. Measurement of Pull-Off Force. We developed the equipment for pull-off force and durability measurements of dry adhesives. The jig with a glass substrate was moved vertically by a motor-driven crank to make contact with a dry adhesive. The dry adhesive was placed on the stage assembled with a z -axis micromanual stage to control the preload, and a micromanual stage that can be tilted was used to adjust the horizontal level. To measure the pull-off force between the dry adhesive and glass substrate, two load cells with a gram-scale resolution and can respond within 100 ms were set below the stage set. The pull-off force was measured with respect to various preloads, and a durability test was conducted at a rate of 50 cycles/min with 40 N preload at room temperature.

2.4. SEM Observation. The fabricated adhesive samples were visually examined by the scanning electron microscopy (SEM) (S-4800, Hitachi), typically at the operating voltage of 10–25 kV after sputtering a thin Au film ($< 5 \text{ nm}$) to avoid electron charging if necessary.

3. Results and Discussion

Figure 1 is a schematic illustration of a continuous production process for dry adhesives consisting of microstructure fabrication and tip inking. Through the whole process, the microstructure was fabricated by UV-assisted molding, the resin to make the adhering part was inked on the tip part of microstructures, and the wide tip was formed by pressurizing with a flat substrate. In the experiment to prove the concept, a 5 cm wide PET film in the roll unit and microstructures mold of 2 cm by 2 cm area were used. In this process, surface treatment of the substrate and the mold is very important. The microstructure was made of PUA and the tip part was made of soft PUA, so that the adhesion

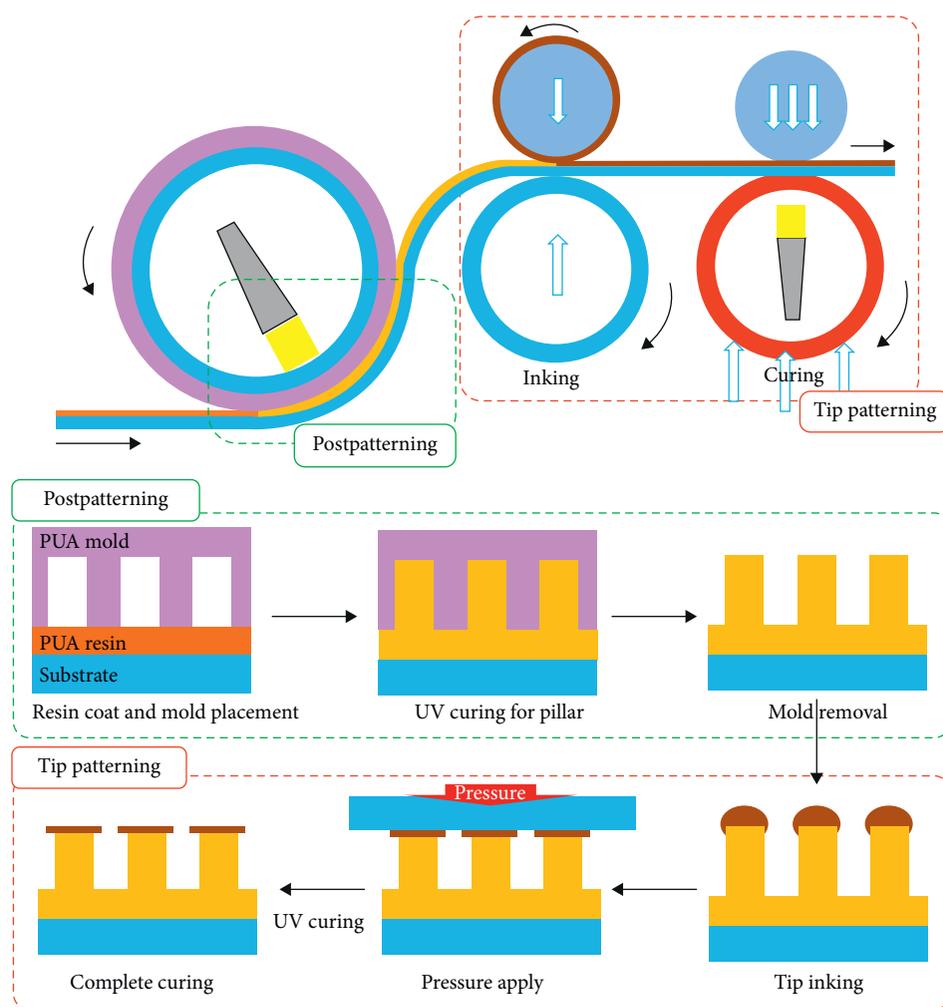


FIGURE 1: Schematic of the continuous fabrication process for mushroom-shaped dry adhesives and procedure for mushroom-shaped structures via imprinting and inking process.

between the two was secured. Because PUA and soft PUA are same kind of polymer-based materials, UV curing has been successfully combined like a single substance. For inking process, a urethane rubber surface was used for proper wetting of the soft PUA, and a PET film with a release treatment was used as the substrate for tip widening. After verifying the possibility of the inking process at the wafer level, the same process principle was applied to achieve continuous production in the roll-to-roll apparatus. The production equipment consists of three process rolls. The first roll is an imprint roll capable of producing a microstructure continuously. We fabricated negative mold of the film type using PUA, and the fabricated soft mold was wound on first roll to realize continuous imprinting. The micropillar structure fabricated on a flexible substrate was then subjected to an inking process through a feeding roll of polymer resin for wide-tip formation. The feeding roll for the inking process was made of urethane and secured with adequate surface energy to transfer the soft PUA to the tip in an appropriate amount. The resin in the tip part is then spread widely by the pressure roll that it contacts. A wide tip was formed on the micropillar by UV exposure with a pressure of about 10 kPa.

The operating speed of the roll unit was determined based on the longest time among the time required for fabricating the microstructure, for inking, and for postexposure. Although not as fast, we were able to produce dry adhesive samples 10 cm long per minute.

Figure 2 shows SEM images of the microstructure of each process of the dry adhesion sample prepared by the aforementioned process. Figure 2(a) is a pillar fabricated by a microstructure production department with a diameter of $10\ \mu\text{m}$, height of $15\ \mu\text{m}$, and period of $35\ \mu\text{m}$. Since the process does not go through an overnight heavy curing process, a reactor still remains on the surface to ensure sufficient wettability and bonding with the soft PUA resin to be inked later. Figures 2(b) and 2(c) are SEM images of the microstructures after inking and tip widening. Due to the sufficient viscosity of the soft PUA, the resin was only present at the tips, and due to the proper wetting it did not flow down the sides of the pillars. In addition, the wide tip was well expressed by the pressure of the PET film roll which was repellent rather than the PUA surface. Although current process conditions have allowed the production of successful dry adhesives, future optimization of the surface energy of the

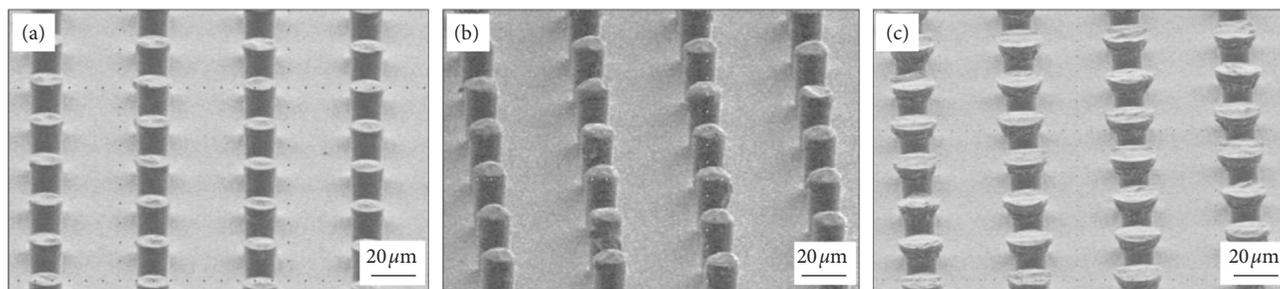


FIGURE 2: SEM images of mushroom structures fabricated in each process: (a) micropillar molding, (b) tip inking, and (c) tip widening.

mold, substrate, and resin will lead to the formation of wider, thinner tips, resulting in high-performance dry adhesives.

Figure 3 shows the measurement results of the pull-off strength, which is a representative performance index of the fabricated dry adhesives. The pull-off strength according to the preload was found to be saturated in the preload of 20 N/cm² or more, and the maximum adhesive property was confirmed as 13 N/cm² in the preload of 40 N/cm². The measured adhesive strength was lower than that of the mushroom-shaped wide-tip microstructures (~20 N/cm²) produced by conventional methods because forming a wide and thin tip necessary for strong adhesion was impossible, which may be solved by optimization of the subsequent process [8].

A comparatively rapid decrease in the adhesive strength was observed during the initial 500 times of use, and the adhesive strength was maintained steadily from about 10 N/cm² to 3000 times of use as shown in Figure 4. After using 1500, 1800, 2100, and 2400 times, cleaning with a Scotch tape was carried out, and the reduced adhesive strength was recovered through this process. Through the cleaning process, the contaminants were removed and the structural collapse, typically pairing, returned to its original state. As the adhesive force recovery through the cleaning process was repeated, the rate at which the adhesive force decreased was gradually increased. Although the adhesion strength is slightly lower than the typical mushroom-shaped microstructure-based dry adhesive, the degradation tendency was considerably better than that of dry adhesives with conventional wide tips. The adhesive strength and lifetime are inversely related to each other; thus, the fabricated dry adhesives are likely to be completely used in applications where they have to be used for a long period of time with a proper adhesive strength. The marathon test result shows a reasonable adhesive strength of up to 3,000 times, and the test is expected to be used in various fields.

4. Conclusions

Demand for dry adhesives has increased in many areas because of their unique performance. However, it was difficult to use because of low yield and long process time of the molding process. In this paper, we have developed a method to continuously fabricate a mushroom-like microstructure by continuously fabricating a simple microstructure using a simple molding process and resin inking. The molded

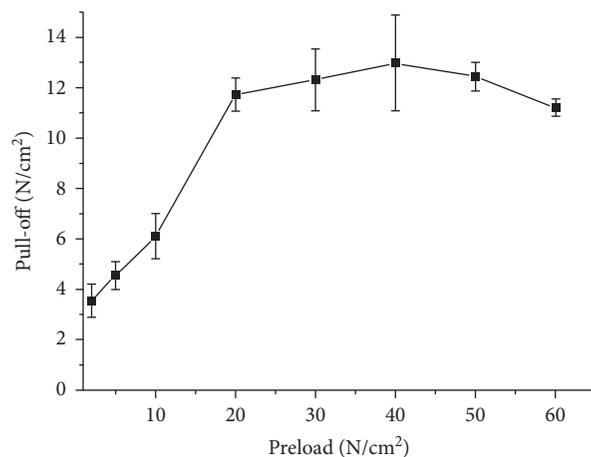


FIGURE 3: Measurement data of the pull-off strength of fabricated dry adhesives as a function of preload.

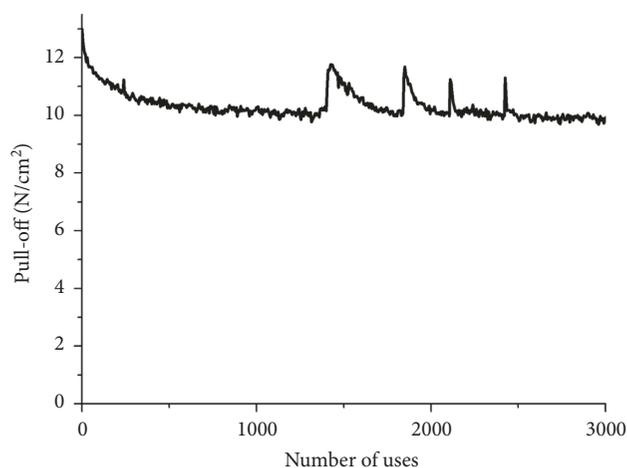


FIGURE 4: Durability test of the fabricated dry adhesives over 3,000 cycles of attachment and detachment.

microstructure acts as a column in the mushroom-like microstructure, and the inked resin acts as a tip through subsequent planarization and curing. The fabricated microstructures showed similar properties to general dry adhesives and showed an adhesion of about 13 N/cm² to the smooth surface. Although the adhesive strength was somewhat lower than that of the conventional dry adhesive production method, the dry adhesive sample produced with

the highly developed technology in terms of productivity did not exhibit a specific adhesive strength drop in 3000 times of use in the marathon test. Further process optimization studies are expected to achieve high adhesive strength when wide tips can be implemented.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

The authors thank Jeong Hyeon Lee for his technical support. MKK acknowledges the support by a grant from the National Research Foundation of Korea (2016R1A2B4007858) funded by the Korean government (MSIP). CWP acknowledges the support by a grant from the National Research Foundation of Korea (2017R1A2B2005515) funded by the Korean government (MSIP).

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