Silicone-based adhesives for long-term skin application: cleaning protocols and their effect on peel strength

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Keywords: adhesives, peel test, reusability, conformal biointerfaces, electronic skin

Abstract

Adhesives that involve adhesion to the skin have been of great technological importance in medical or pharmaceutical fields, including recently emerging wearable sensors and electronics. The objective of this work was to evaluate the performances of silicone-based adhesives with skin, using a peel adhesion test. Specifically, we explored the effect of adhesive cleansing, which is an inevitable daily event for patients’ comfort in long-term applications. Firstly, three medical grade silicone gels, Silbione® RT 4717, Silbione® RT 4642, and Silpuran® 2130, were used to fabricate adhesive pads. Their peel strength values were subsequently measured and compared, among which Silbione® RT gel 4717 possessed the highest peel strength. Therefore, it was selected as the raw material to fabricate the pads with a thickness range of 640–740 μm. Secondly, the peel adhesion of Silbione® RT 4717 adhesive pad was further compared with a series of commercial products that employ various medical-grade adhesives. The peel strength results indicated that our custom-made adhesive pad had an adequately strong adhesion for clinical use. Thirdly, in order to observe and predict the long-term performance of the adhesives, an aging test was performed in an ambient environment, revealing that Silbione® RT 4717 adhesive remained highly sticky for 5 days. Lastly, adequate cleansing protocols were established by monitoring the changes in peel strength after washing and wiping events. The reusability analysis showed that Silbione® 4717 adhesive pad was reusable in a one-week period for the washing method and 3 days for the wiping method.

1. Introduction

Adhesives that involve adhesion to the skin have been widely applied in a variety of medical or pharmaceutical fields, including the securing of medical devices [1–4]. For this reason, adhesives are critical to the success of bonding dressings or medical devices that require intimate contacts of dressings or electrodes with the skin surface [5]. Recent advances in skin-adhesive diagnostic devices accompany increased weight and complexity of the medical devices [6–10], which necessitate research on long-term reusable skin adhesives and their cleansing protocols. For such skin application, an adhesive must have sufficient initial tack to secure the position of the device upon application, and must provide enough stickiness to hold the device in the correct position throughout the wearing time. Furthermore, the removal of these adhesives should not cause trauma to the skin. These adhesives are also required to be non-irritant, non-sensitizing and residue-free [11].

Adhesives exhibit a viscoelastic form and develop adhesion to the skin and cohesion to resist debonding [12]. The three most commonly used methods to monitor adhesive performance are tack, shear strength, and peel adhesion tests [4, 13, 14]. Tack refers to an instantaneous level of adherence of an adhesive. It is a short-term measurement of skin adhesion, which only demonstrates the initial tendency to stick to the skin. In order to hold adhesives in position,
a high level of tack is essential. Shear strength, also known as cohesion, reflects the internal or cohesive strength of the adhesive rather than the bond between the adhesive and a substrate. It describes the ability of an adhesive to resist shearing forces. Peel adhesion is determined by the ability of adhesives to resist separation from the skin surface after a period of application. It is complex, multifaceted, and broadly applicable from cell adhesion to bio-fouling, thus is one of the most ubiquitous phenomena in nature and technology [15]. Upon initial application to the skin, adhesion is at its maximum, but gradually decreases due to transdermal water evaporation and skin desquamation [11].

Peeling is the most commonly observed mode of adhesive bond failure [16]. The peel force or peel adhesion, which is applied to rupture the adhesive bond [17], is often used as an index of bond quality. This force can be quantified by performing a controlled peel test that is conducted by pulling the adhesive at a constant rate from a known substrate [16]. Many factors have been reported to influence peel force, including peel rate, peel angle, contact pressure, property of the substrate, withdrawal speed, temperature, and humidity of the environment [18]. However, peel force is not representative of the true strength of the adhesive bond. The measured peel force is a combination of the true strength of the adhesive bond and the energy dissipated due to the elastic and plastic deformation of the adherends [19]. These mechanics would be of great help in the interpretation of test results on the failure mechanism of the bond, especially when the substrate material is elastic instead of rigid [16].

Obtaining peel test data on human skin is highly complex primarily due to the great variability of skin and its properties as well as the difficulty of executing realistic experimental set ups [20]. To simplify this, most standard peel tests adopt flat and rigid substrates, such as steel and glass. The results from these surfaces are precise and reproducible, but the findings are not easily related to testing on human skin [21]. One of the major difference is the rough contour of human skin, which reduces the effective area of intimate contact between the adhesive and the skin, leading to a reduced adhesion [20]. Several studies [20, 22] have proposed the use of synthetic substrates which mimic the mechanical and surface properties of human skin, however, such synthetic films are unable to reproduce the rheological behavior of skin and present with different modes of failure [23]. Considering these difficulties, pig skin has been suggested to be a suitable model to replace human skin during peel test. The anatomy, topical features such as texture and roughness ($R_s$ values, the arithmetical mean value of the amounts of the ordinate value within an individual measuring distance, for both human and pig skin are $20 \pm 3 \mu m$ [24]), and the biochemical property of pig skin are remarkably similar to those of human skin [21]. Therefore, hair-removed pig skins have become increasingly utilized as a substrate material to mimic human skin for topical adhesion studies [25, 26].

In addition to having the appropriate peel strength, adhesives designed for human skin must also be sticky, comfortable to wear, non-irritant and residue-free. The three most commonly used commercial adhesive materials today are rubbers, acrylics and silicones. Natural rubber adhesives prevailed in the market until 1960s, but their high cost, the shortage of supply, and the advances in polymer chemistry rendered the dominance to synthetic materials [1]. Traditionally, acrylate-based adhesives have been widely used for skin contact applications due to their inherent tack, strong oxidative stability, and easy adaptability to a variety of needs [4]. However, nowadays adhesives for medical applications are called upon to meet a multitude of requirements including less skin irritation and better reusability. The silicone adhesives, which possess several potential advantages, would be an ideal solution for prolonged use of the dressing and electronic patch. First, the silicone adhesives’ viscoelastic flow characteristics can create large intimate contact area on the skin, where the surface topography is extremely uneven. Second, their tackiness may not decay or increase over time, whereas the strength of adhesion is adequate for skin application [11]. Third, silicone adhesives are residue-free and relatively inert with minimum risk of skin intolerance [27–29]. Fourth, silicone adhesives have significantly higher moisture vapor transmission rate and fluid handling capacity than other adhesives, which are crucial factors that govern patients’ comfort during long-term applications [30].

In this study, we fabricated a series of silicone-based adhesive pads and evaluated their performances using a peel test. The peel strength, a measure of the minimum force required for the delamination to occur [31], was quantitatively determined on the cadaveric pig skin. Specifically, we targeted the use for surface electromyography electrode pads, where reusability after frequent washing is critical for patients’ comfort [32, 33]. For this objective, an adequate washing protocol must be established by monitoring the change in peel strength after washing events. Here, we evaluated three types of commercially available silicone gel adhesives. For the silicone gel with the highest peel strength, the optimal thickness that balances between the peel strength and the patients’ comfort (no residual adhesive upon removal) was identified. The peel strength of the silicone gel at the optimal thickness was compared with a series of over-the-counter medical-grade adhesives. An aging test also was performed to observe and predict the long-term performance of the adhesives. For the reusability study, we quantitatively evaluated washing and wiping protocols to assess if prolonged use of the adhesive is possible. In addition, washing tests and wiping tests were compared to determine a better cleaning method for patients.
Table 1. The recommended applications and published peel strength of three types of silicone gels tested in our study (from their product brochures).

| Commercial name of silicone gel | Manufacturer        | Recommended applications                  | Peel strength (N mm⁻¹) |
|--------------------------------|---------------------|-------------------------------------------|------------------------|
| Silbione® RT 4717              | Bluestar Silicons   | Adhesive wound dressing                    | 0.036                 |
| Silbione® RT 4642              | Bluestar Silicons   | Adhesive sheeting for scar treatment       | 0.046                 |
| Silpuran® 2130                 | Wacker Chemie AG    | Skin adhesive                              | 0.072                 |

¹ 180° peel using Bristol paper.
² Peel test on stainless steel.

2. Materials and methods

2.1. Materials and fabrication of adhesive pads

Silbione® RT 4717 and Silbione® RT 4642 were supplied by Bluestar Silicons (New Jersey, USA). Silpuran® 2130 was provided by Wacker Chemie AG (München, Germany). For peel test, a commercial fabric that does not allow dimensional change during the test was used as a backing substrate. Hair-removed pig skin was purchased from a local grocery store in Edmonton, Canada and was used as a model substrate for peel test. Sunlight® detergent and standard isopropyl alcohol swabs were obtained from Shoppers Drug Mart (Edmonton, Canada). Dynarex® Clear Surgical Tape (Dynarex, New York, USA), Nexcare® Sensitive Skin Tape (3M, Minnesota, USA), Band-Aid® (Johnson & Johnson, New Jersey, USA), Easy@home Re-useable TENS & EMS carbon electrode pad, and original Acrylic Pad were purchased from Amazon in Canada.

Three types of silicone gel, Silbione® RT 4717, Silbione® RT 4642, and Silpuran® 2130, were used to fabricate adhesive pads in this work. Their recommended applications and published peel strength values on rigid subjects were summarized in table 1; these values were adopted from their respective product brochures (technical data sheets provided by Bluestar Silicons and Wacker Chemie AG). General fabrication procedures were as follows. First, the fabric backing substrate was cut into a rectangular shape with a dimension of 50 mm × 30 mm by an automatic cutting machine (ScanNcut, Brother Industries, Japan). Next, the two parts (referred to parts A and B in this paper) of the silicone gels were thoroughly mixed by hand with a 1:1 weight ratio. The mixture was then placed in a vacuum chamber to degas for 30 min to eliminate any entrapped air. After the adhesive was degassed, between 1 and 1.2 g of the mixture was spread evenly on the fabric backing material and left at the room temperature of 21 °C for 1 d to be cured. All adhesive pads were covered in petri dishes. The target thickness of the adhesive pad was in the range of 640–740 μm, and was verified by a caliper measurement. The interval between the manufacturing and the measurement was controlled to be 24 h.

Two additional thickness levels of adhesive pads using Silbione® RT 4717 were prepared. To do so, 0.6–0.7 g and 2–2.2 g gel mixtures were used to obtain adhesive layers with thicknesses of 240–340 μm and 1240–1340 μm, respectively. These pads were manufactured to be compared with the Silbione® RT 4717 pads with a thickness range of 640–740 μm. Thus, an optimal thickness for Silbione® RT 4717 adhesive pads could be selected.

2.2. Peel strength analysis of adhesive pads with Silbione® RT 4717, Silbione® RT 4642, and Silpuran® 2130 (Study 1)

A 90° peel test was conducted on pig skin to determine the peel strength of the fabricated adhesive pads with Silbione® RT 4717, Silbione® RT 4642, and Silpuran® 2130 using the Instron 5943 with a 1 kN load cell. The roughness value of pig skin was measured by a Zygo optical profilometer (Zygo Corporation, United States) and $R_m$ was 6.191 mm. The pig skin was secured to a platform, and the adhesive pad was then placed on the pig skin by pressing with the index finger. The average peel strength of index finger is reported to be 41.6 N from 100 subjects (50 male and 50 female) with the age between 18 and 29 years old [34]. The adhesive pad was attached with a piece of sticky tape, which was secured in a set of upper clamps. The adhesive pad was peeled mechanically from the pig skin at a speed of 10 mm min⁻¹. The average peel strength (N mm⁻¹) was calculated by measuring the average load (N) of the peel test and dividing it by the width (mm) of the bonded pad [35] (figure 1). Each test was repeated five times to evaluate the average peel strength.

Two independent samples t-tests were used to compare peel strength between the Silbione® RT 4717 and two other adhesive pads: Silbione® RT 4642 and Silpuran® 2130 adhesive pads. A Bonferroni adjusted alpha for two comparisons was used ($\alpha = 0.025$).

2.3. Thickness test of the Silbione® RT 4717 adhesive pads (Study 2)

Peel tests were performed on Silbione® RT 4717 adhesive pads with three thickness ranges: 240–340 μm, 640–740 μm, and 1240–1340 μm. The same procedures were used to quantify the effects of differences in
thickness on the peel strength. Three independent samples $t$-tests were used to identify differences in peel strength between the different thickness ranges. Three comparisons were made and a Bonferroni adjusted alpha was applied here ($\alpha = 0.017$).

2.4. Peel strength comparison of Silbione® RT 4717 and various commercial medical-grade adhesives (Study 3)

Peel test was conducted to determine peel strength values of commercial adhesive pads—Dynarex® Clear Surgical Tape, Nexcare® Sensitive Skin Tape, Band-Aid®, Easy@home Re-useable TENS & EMS carbon electrode pads, and Acrylic Pad—by following the same procedures in section 2.2. Afterwards, comparisons of peel strength values were made between Silbione® RT 4717 and these commercial medical-grade adhesives. An independent samples $t$-test was used to compare peel strength between the Silbione® RT 4717 adhesive and Easy@home Re-useable TENS & EMS carbon electrode pad ($\alpha = 0.05$).

2.5. Aging test of the Silbione® RT 4717 adhesive pads (Study 4)

Silbione® RT 4717 adhesive pads with thickness of 640–740 $\mu$m were chosen to perform an aging test over a 7 d period according to our previous testing results. Three adhesive pads were kept in petri dishes with the adhesive side facing up at room temperature (21 °C) for 7 d. The pads were removed from the petri dish once a day and subjected to a peel test following the same procedures described in the peel strength analysis. A control group was also included in this experiment. All adhesives pads were enclosed in the petri dishes, thus they are not exposed to the outside environment. An independent samples $t$-test was performed to compare peel strength between days 5 and 6. The difference in day 6 between experimental and control groups was also evaluated by an independent samples $t$-test.

2.6. Microscopic examination of the Silbione® RT 4717 adhesive pads (Study 5)

The surface of Silbione® RT 4717 adhesive pads after attached to human skin was observed by scanning electron microscope (SEM) (Zeiss EVO MA10, Zeiss, Germany). The freshly prepared adhesive surface was used as a control group. Samples were mounted onto aluminum stubs and sputter-coated with gold for SEM examinations. The fouling surface was compared to the freshly made surface to study adhesives’ properties.

2.7. Cleansing protocols and reusability tests of the Silbione® RT 4717 adhesive pads (Study 6)

Reusability of the Silbione® RT 4717 adhesive pads with the thickness between 640 and 740 $\mu$m was evaluated by two cleansing protocols: washing test and wiping test.

Washing Test: Each pad was washed in 50 ml warm water (35 °C) mixed with 5 ml dish detergent to remove residue from the adhesive after each attachment to the pig skin. Four different washing times of 5, 10, 20, and 40 s were tested. After each wash, the adhesive pads were dried by compressive air for 20 s. For each washing condition, peel tests were performed after 1, 5, 10, and 20 attach/detach cycles.

The washing test data were analyzed by a two-way analysis of variance (ANOVA) with two independent variables (IV): 1 IV with 4 levels (wash times with 5, 10, 20, and 40 s) and 1 IV with another 4 levels (attach/detach cycle with 1, 5, 10, and 20 cycles). The ANOVA was used to determine effects of these two IVs on the peel strength of the Silbione® RT 4717 adhesive pads.
If a main effect for IV was found, pairwise comparisons between all levels were conducted.

Wiping Test: A similar test was completed where an adhesive pad was cleaned with a standard isopropyl alcohol swab. Four different conditions, each with a different number of swab strokes were compared. The conditions were: zero wipes (no cleaning), 1 wipe, 5 wipes, and 10 wipes per one attach/detach cycle. For each alcohol swab wipe number, peel tests were performed after 1, 5, 10, and 20 attach/detach cycles.

Similarly, a two-way ANOVA was performed to determine the effect of various numbers of alcohol swabs and attach/detach cycles on adhesives’ peel strength. The two IVs were: number of alcohol swabs (zero wipe, 1 wipe, 5 wipes, and 10 wipes) and number of attach/detach cycles (1, 5, 10, and 20 cycles). In addition, two independent samples t-tests were planned to compare peel strength for 10 cycles and 5 cycles when 1 wipe and 5 wipes were used. A p value less than 0.05 was considered statistically significant for both 1 wipe and 5 wipes conditions.

A control group experiment (no cleansing after each attach/detach cycle) was conducted to evaluate the efficacy of the cleansing method.

3. Results

3.1. Study 1. Peel strength analysis of adhesive pads with Silbione® RT 4717, Silbione® RT 4642, and Silpuran® 2130

The mean peel strength values of Silbione® RT 4717, Silbione® RT 4642, and Silpuran® 2130 adhesive pads with the thickness between 640 and 740 μm were measured on the pig skin and summarized in figure 2. The results revealed that Silbione® RT 4717 has the maximum peel strength of 0.014 N mm⁻¹, which is obviously higher than those of Silbione® RT 4642 and Silpuran® 2130 adhesive pads. Therefore, the Silbione® RT 4717 gel was selected as the raw material to fabricate adhesive pads. An independent samples t-test was performed and indicated statistically significant differences (∧ = 0.008, α = 0.025) in the mean peel strength for Silbione® RT 4717 and Silbione® RT 4642. Similarly, the mean peel strength comparison between Silbione® RT 4717 and Silpuran® 2130 was also significantly different (∧ = 0.007, α = 0.025).

3.2. Study 2. Thickness test of the Silbione® RT 4717 adhesive pads

The peel strength values of Silbione® RT 4717 adhesive pads with three thickness ranges from 240–340 μm to 640–740 μm and finally to 1240–1340 μm were compared in figure 3. It could be observed that the peel strength increased as the adhesive layer became thicker. The result of an independent samples t-test was statistically significant for the comparison group with thickness range of 240–340 μm and 1240–1340 μm (∧ = 0.016, α = 0.017). However, the independent samples t-test results were not statistically significant for the two other comparison groups: 240–340 μm and 640–740 μm (∧ = 0.16, α = 0.017); 640–740 μm and 1240–1340 μm (∧ = 0.29, α = 0.017). It is notable that the 1240–1340 μm samples tended to leave residuals when peeled off, whereas the 640–740 μm sample removed cleanly.

3.3. Study 3. Peel strength comparison of Silbione® RT 4717 and various commercial medical-grade adhesives

The peel strength of Silbione® RT 4717 was compared with a series of commercially available medical-grade adhesives. The result illustrated in figure 4 showed that Silbione® RT 4717 adhesive pad exhibited a substantially higher peel strength especially when compared to Dynarex® Clear Surgical Tape, Nexcare® Sensitive Skin

Figure 2. Peel strength of adhesive pads with three different commercial medical-grade silicones at the thickness range between 600 and 800 μm. The error bars represent the standard deviation.
Tape, Band-Aid®, and Acrylic Pad. The peel strength of Silbione® RT 4717 adhesive pad was slightly higher than that of the carbon electrode (Easy@home®) pad based on the observation from figure 4. An independent samples t-test displayed no statistically significant differences in the mean peel strength for Silbione® RT 4717 adhesive pad and carbon electrode (Easy@home®) pad ($p = 0.50, \alpha = 0.05$).

### 3.4. Study 4. Aging test of the Silbione® RT 4717 adhesive pads

The degradation in peel strength of adhesives was tracked over the course of one week [19] and displayed in figure 5. It showed that the peel strength increased on day 2 compared to that of day 1 and then decreased but remained stable between days 3 and 5. On the sixth day, the peel strength suddenly dropped to around 20% of the original strength. When analyzed by an independent samples t-test, a statistically significant difference ($p = 0.05$) in the mean peel strength between days 5 and 6 was found. In the control group, peel strength values of adhesive pads were basically invariable within a week. Throughout the study, peel strength values of adhesives in the experiment group were consistent with those of the control group from days 1 to 5, but great differences occurred after day 6 (however, $p = 0.10$ was still not significant in independent t-test for day 6).

### 3.5. Study 5. Microscopic examination of the Silbione® RT 4717 adhesive pads

Figure 6 presented the SEM micrographs of the adhesives that are freshly made (figure 6(a)) and after an attach/detach cycle (figure 6(b)). The control group
of freshly prepared adhesive pad, displayed a relatively smooth and clean surface. However, after the adhesive pad was attached to human skin and then detached, a rough surface occurred with hints of debris and fibrous materials.

3.6. Study 6. Cleansing protocols and reusability tests of the Silbione® RT 4717 adhesive pads

Figure 7 showed results from washing reusability tests. Two main effects, washing time and attach/detach cycles, were investigated here to study their effects on peel strength of adhesives.

First, it can be observed that washing time had an impact on peel strength as 5 s wash displayed obviously lower peel strength values than those of three other washing times under 1, 5, and 10 attach/detach cycles. This was confirmed by a two-way ANOVA, which indicated a statistically significant difference ($p < 0.001$) for washing time comparisons. As for attach/detach cycles, peel strength values after 20 cycles were significantly lower ($p = 0.010$) than those after 1, 5, 10 cycles. This result applied to adhesive pads under all 4 washing times.

To further investigate the effects of washing time and attach/detach cycles on peel strength, different comparisons were carried out. When we compare the washing times, 20 s wash showed the highest peel strength, but the differences from 10 to 40 s washes are less than obvious; a two-way ANOVA did not show a statistically significant difference ($p = 0.314$) between 10 and 40 s wash. When we compare the washing cycles, a two-way ANOVA did not show statistically significant differences in peel strength when compared 10 cycles to 1 cycle ($p = 0.37$) and 5 cycles ($p = 0.25$). After 20 cycles, however, the peel strength for 10 s wash decreased to 30.57% of the original strength; for 5 s, 20 s and 40 s, the numbers of remained peel strength were 51.78%, 59.85% and 48.6%, respectively. Peel strength of adhesives without wash cleaning (no wash) was 18.9% to the original strength,
which was the lowest among all washing conditions. The result of two-way ANOVA was statistically significant as 20 cycles was compared to 10 cycles ($p = 0.020$), 5 cycles ($p = 0.001$), and 1 cycle ($p = 0.002$), which agreed with our observations.

Wiping test in figure 8 was another technique to study the reusability of adhesives. Similarly, both alcohol swabbing numbers and attach/detach cycles had an impact on the adhesives’ adhesion strength. The results of a two-way ANOVA were statistically significant for the wiping number ($p = 0.01$) and cycles ($p = 0.004$). For both 1 wipe and 10 wipes cases, adhesives showed significantly higher rate of decay after 5 attach/detach cycles from 1 cycle results. The peel strength of 1 wipe at 1 cycle should be an outlier here as its value was higher than 0.014 N mm$^{-1}$, which was the original peel strength of Silbione® 4717 adhesives. The result of 5 wipes at 1 cycle seems to be an outlier as well because it showed an inconsistently low value. The two results may be experimental errors. All wiped samples (0, 1, 5, and 10 wipe(s)) showed similar peel strength values in 5, 10, and 20 cycles. After 20 cycles, the peel strength of 0, 1, 5, 10 wipe(s) case were 18.9%, 39.1%, 49.6% and 51.2% to the original peel strength, respectively.

4. Discussion

Silicone-based adhesives consist of two basic components, the polydimethylsiloxane with a high glass transition temperature ($T_g$) and the silicate resin with a low $T_g$. The adhesive properties of silicones are
dependent on the ratio of polydimethylsiloxane to resin, as high polymer content generates a soft but tacky adhesive, whereas high resin content produces a non-sticky but strong adhesive [15]. Three silicone gels used in this work, Silbione® RT 4717, Silbione® RT 4642, and Silpuran® 2130, are two-part platinum catalyzed elastomers that crosslink at the room temperature by polyaddition reaction. In general, part A contains a vinyl functional polymer and the platinum catalyst, while part B contains the hydride crosslinker and inhibitor. The polyaddition cure is the most common cure mechanism to prepare medical-grade silicone elastomers [36]. Catalyzed by platinum, the hydride and vinyl functionalities would react to generate a cured silicone elastomer.

Human skin has a complex texture with viscoelasticity; pig skin resembles these properties, thus we performed peel test on pig skins [37]. In figure 2, the mean strength for Silbione® RT 4717 was greater than those of Silbione® RT 4642 and Silpuran® 2130, and statistic results were significant for the two comparisons (Silbione® RT 4717 and Silbione® RT 4642; Silbione® RT 4717 and Silpuran® 2130). Therefore, Silbione® RT 4717 gel was selected as the raw material to fabricate the adhesive pads for our further investigations. It was also noteworthy that the trend of the peel strength values provided by figure 2 was different from those listed in table 1. This could be attributed to the variations of interactions between these three silicone gels and the skin.

The adhesive thickness has an important effect on the peel adhesion property of adhesives. Figure 3 showed that peel strength of Silbione® RT 4717 adhesives increased with increasing thickness of silicone gel. This was because the bonding strengthens as the adhesive gel becomes thicker. However, it was reported that after a certain value of coating thickness, peel adhesion would remain stable or even decrease with the addition of thickness, whereas the threshold for the constant adhesion strength depends on the combination of adhesive and substrate [19]. This can be attributed to the shift from cohesive to adhesive failure at the threshold [38]. But in our study peel strength increased monotonically as adhesive thickness increased, which indicates that our thickness is below the threshold thickness. The adhesives with a thickness range of 640–740 μm exhibited higher peel strength compared to the adhesives with that of 240–340 μm; meanwhile it was not being too thick to leave residues on the skin. As a result, 640–740 μm was selected as the best thickness of the adhesive layer, as evidenced by appropriate peel strength (compared to other commercial medical-grade adhesives) and residue-free property.

To further evaluate the peel adhesion of Silbione® RT 4717 adhesive pads for practical application, its peel strength was compared with a series of commercially available medical-grade adhesives. Dynarex® Clear Surgical Tape, Nexcare® Sensitive Skin Tape, Band-Aid®, and Acrylic Pad are adhesives coated with a thin layer of silicone or acrylic gel, thus possessing relatively low stickiness and reusability. However, Easy@home Re-useable TENS & EMS carbon electrode pads are made of high-quality reusable carbon adhesive gel with high tackiness and good reusability. According to the comparison results in figure 4, the peel strength of Silbione® RT 4717 adhesive pad was quite comparable to that of the Easy@home Re-useable adhesive pad, indicating that the custom-made adhesive pad had an adequately strong adhesion for clinical use.

The adhesive performance is susceptible to environmental aging during the wearing period on the skin [39]. All adhesives exhibit strong initial tackiness, but their peel adhesion degrades over time and embrittlement occurs gradually. It arises from the chain scission and increased crosslinking of adhesives, which are caused by thermos-oxidative degradation and hydrolysis [40]. For long-term therapy, it is reasonable to predict a scenario that requires the application and removal of an adhesive pad during a one-week-long duration. This scenario requires high demand on the long-term performance of adhesive. In order to observe and predict such long-term performance for the Silbione® RT 4717 adhesive pad when the silicone is not protected by a protecting layer (in normal storage condition, the adhesive products are typically protected by a coated paper), an aging test was performed in an ambient environment. A control group was also carried out to study the environmental effects on peel adhesion where all adhesives were enclosed in petri dishes throughout the whole test process. Figure 5 displayed the degradation in peel strength of adhesives over a-week time. From days 1 to 5 peel strength values of adhesives in the experiment group were almost the same with those of the control group, but after day 6 they were greatly lower. The strength of adhesive bond degraded due to the presence of water and oxygen. The polar nature of water molecules could disrupt the secondary bonds across interfaces, resulting in the adhesive delamination [41]. Oxygen affected the strength because it oxidized the adhesive. From the results we can conclude that the Silbione® RT 4717 adhesive was highly sticky for 5 d with nearly constant peel strength; however, the adhesion strength strongly dropped after 5 d, thus longer storage without the protecting layer is not recommended.

Reusability is arguably the most important concern for skin adhesives for most practical applications. In the bonding process, an optimal adhesive is required to flow and deform easily to contact the surface conformably. During the peeling process, on the other hand, it should provide sufficient cohesive strength to prevent a bulk failure, which results in residue formation on skin. In addition, a large energy dissipation is needed so that a greater work of adhesion can be achieved. The peeling process is always accompanied by morphological changes such as fingering, occurrence of a rough surface and particulate
contamination, which render the adhesive to lose its adhesion force [42]. Therefore, the fouling of surface is another important factor that limits the reusability of adhesives. To have a better understanding on this effect, a SEM analysis was performed on adhesive surface and some contaminates were observed to be human hairs and dead skins.

From the above SEM analysis, it was found that adhesive surface was contaminated after applied onto skin, which greatly limited the adhesives’ reusability. To overcome this disadvantage, an efficient cleansing protocol is necessary. In this study, the reusability of Silbione® RT 4717 adhesives were investigated by warm soapy water washing method and isopropyl alcohol wiping method. Specifically, washing test and wiping test were compared to propose a better cleaning method for patients.

In the washing test, the peel test was repeated 3 times after 1, 5, 10, and 20 attach/detach cycles for each washing time to determine the optimal washing time as well as the reusability characteristics. Results showed that both washing time and attach/detach cycles had effects on adhesive’s peel strength. Among all washing conditions, 5 s wash displayed the lowest peel strength at 1, 5, and 10 attach/detach cycles. This could be explained by that 5 s wash did not provide enough time to remove the debris or dirt attached onto silicone gels. Consequently, effective contact area between the adhesive pad and skin decreased, leading to weakened interface bonding. After 20 cycles, the 20 s wash retained the most peel adhesion. One possible reason is that when the washing time was less than 20 s, adhesives pads could not be cleaned sufficiently by warm soapy water; whereas if the washing time goes up to 40 s, the soapy water might have some negative effects to the adhesion of silicone. After 20 cycles the peel strength decreased to the range between 48% and 60% of the original strength. Decreasing adhesion strength may be owing to the generation of a rough surface that occurred during repeated attachment and detachment to pig skin. Surface contaminations by residual dirt and debris that were not cleaned after each cleaning cycle was another factor that limited adhesives’ reusability [42]. If these adhesive pads were cleaned three times a day with soapy water washing, and the 50% of original peel strength (still higher than that of Band-aid®) was necessary for the application, 7 d (20 attach/detach cycles) would be the suggested lifetime of the product; 20 s would be the recommended washing time as it retained the most peel strength. Combining the factor of aging effect, we can conclude that the adhesive pad was reusable in 5 d, and cleaning was possible. It is notable that the actual application on the skin of living human has additional issues, such as shedding of dead cells and accumulation of sweat.

In the wiping test, for 1 wipe and 10 wipes cases, samples showed a high decay after 5 attach/detach cycles. Similar to the washing test, reduced adhesion strength could be attributed to dirt and debris, but it is also possible that the abrasion from swabbing process caused the physical damage of the adhesives layer. In addition, isopropyl alcohol that did not evaporate completely may swell the silicone adhesive leading to a loss in physical properties. Adhesive strength seems to stabilize after 5 attach/detach cycles. After 20 cycles, 10 wipes show higher adhesion strength than 0 and 1 wipes. 50% of the original peel strength is still higher than that of Band-aid® and could be used for practical applications. Here, our result shows that the adhesion strength was successfully maintained after 20 cycles of attach/detach cycles followed by 5 and 10 wipes by isopropyl alcohol swab. If the adhesive pads were to be cleaned three times a day, our result suggests that our adhesive can be used at least for 7 d. Combining the factor of aging effect, 5 d would be the suggested lifetime of the Silbione® RT 4717 adhesives.

When comparing the soapy water washing with the isopropyl alcohol wiping, it was found that the water washing method retained the adhesion strength better during the cleansing process. This might be owing to the mild property of soapy water. Therefore, we would advise using warm soapy water washing for 5 d to clean the prepared Silbione® RT 4717 adhesive pads.

5. Conclusions

In this work, three types of silicone-based adhesive pad were fabricated and evaluated by peel tests on the pig skin. Among them, Silbione® RT 4717 gel was selected as the raw material to fabricate the adhesive pad due to its relatively higher peel strength values. The thickness range of 640–740 μm was sufficient to provide appropriate adhesion, meanwhile it was deemed not too thick to leave residues on the skin. Therefore, it was a suitable thickness range for Silbione® RT 4717 adhesive pads. The stickiness of Silbione® RT 4717 adhesive pad with thickness range of 640–740 μm was further evaluated by comparing to commercially available adhesives. Results indicated that our custom-made adhesive pad had an adequately strong adhesion for clinical use. In addition, an aging test showed that Silbione® adhesive remained highly sticky for 5 d with nearly constant peel strength when exposed to ambient atmosphere, whereas prolonged exposure to water and oxygen degraded the peel strength after 5 d of exposure. Reusability of the adhesives with repeated cleansing cycles also was evaluated by washing and wiping tests; results revealed that, under standard cleansing frequency, Silbione® 4717 adhesive pad was reusable in 5 d period for both washing and wiping methods.

Acknowledgments

We would like to thank Shide Qiu and Chungyeon Cho for the help with SEM analysis. We are grateful for Alberta Cancer Foundation (Transformative Program) and CIHR/NSERC (Collaborative Health Research Projects) for funding. We also appreciate
Professor A. L. Elias for the use of Instron peel tester with support from Canada Foundation for Innovation (CFI) Leader’s Opportunity Fund.

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