Appl. Chem. Eng., Vol. 35, No. 4, August 2024, 341-345
https://doi.org/10.14478/ace.2024.1040

Article

바질 씨앗을 이용한 식용·친환경 접착제 개발

이현윤 · 이혜주 · 류성호* · Daniel Sim** · 김대석†

부경대학교 고분자공학전공, *한국건설생활환경시험연구원 고분자소재센터,
**Integrative Health & Performance Sciences Division, UES
(2024년 7월 5일 접수, 2024년 7월 22일 수정, 2024년 7월 23일 채택)

Development of Eco-friendly and Edible Adhesive Using Basil Seed Mucilage

Hyun Yun Lee, Hye Joo Lee, Seong Ho Ryu*, Daniel Sim** and Dae Seok Kim†

Department of Polymer Engineering, Pukyong National University, Busan 48513, Korea
**Applied Polymer Research Center, Korea Conformity Laboratories, Incheon 21594, Korea*
***Integrative Health & Performance Sciences Division, UES, Inc, Dayton, OH, 45432 USA*
(Received July 5, 2024; Revised July 22, 2024; Accepted July 23, 2024)

초 록

일반적인 접착제에는 인간과 환경에 유해한 화학물질이 포함되어 있어 안전 문제가 발생할 수 있다. 그러나 활용되지 않는 자원 중 하나인 바질 씨에서 해결책을 찾을 수 있다. 우리는 바질 씨에서 점증 물질을 추출하고, 바질 씨 점질물(BSM)과 물의 비율을 조절함으로써 접착력을 통제할 수 있다는 사실을 발견했다. 물이 증발하고 BSM 농도가 증가함에 따라 접착력이 향상되었다. BSM은 접착제뿐만 아니라 점착제로도 사용될 수 있다. 이 연구는 BSM의 인상 깊은 강도와 안정성을 선보여, 이를 이용한 산업용 친환경 접착제가 식물 기반 자원에서 나온 혁신적인 선택지임을 입증한다.

Abstract

Common adhesives contain harmful chemicals, posing risks to humans and the environment. Basil seeds, an underutilized resource, can offer a solution. We extracted mucilage from basil seeds and found that altering the basil seed mucilage (BSM) to water ratio allowed us to control the adhesive strength. As the concentration of BSM increased and the water evaporated, adhesive strength improved. BSM can serve as both an adhesive and pressure-sensitive adhesive. This research showcases BSM's impressive strength and stability, making it a promising eco-friendly industrial adhesive option from plant-based sources, revolutionizing the adhesive industry.

Keywords: *Basil seed mucilage, Edible adhesive, Tackifier, Eco-friendly*

1. Introduction

From an industrial perspective, the adhesives utilized in residential construction, which can directly impact respiratory health, are currently the focus of intensified efforts in developing environmentally friendly alternatives to mitigate their adverse effects. Adhesives that consider terrestrial and atmospheric ecosystems are gaining significant attention as a potential means of reducing chemical usage and promoting environmental sustainability[1].

Basil (*Ocimum basilicum L.*) belongs to the *Lamiaceae* family and is extensively cultivated in the Himalayan region of India, particularly in the Kashmir valley. The seeds of this plant produce mucilage when immersed in an aqueous solution[2]. The resulting mucilage firmly adheres to the seed core, playing a pivotal role as a substantial source of polysaccharides and soluble fibers. The quantity of mucilage that can be extracted from basil seeds is approximately 20.5% (w/w). The incorporation of such hydrocolloids into various foods imparts multiple functional properties, including oil absorption capacity, water retention, swelling capacity, foam formation capability, and emulsification. Therefore, it is imperative to comprehensively analyze the functional, rheological, and morphological characteristics to formulate and implement a strategy for employing basil seed mucilage as tackifiers or adhesives[3,4].

Basil, recognized for its medicinal applications in treating renal dysfunction-related conditions such as headaches, coughs, and diarrhea, as well as its utilization as a food and additive in pharmaceutical and cosmetic products, serves as the foundational component for our research. We will produce adhesives by evaporating the solvent, water, while also developing tackifiers by incorporating glycerol to retain the solvent.

† Corresponding Author: Pukyong National University
Department of Polymer Engineering, Busan 48513, Korea
Tel: +82-51-629-6448 e-mail: daeseok@pknu.ac.kr

pISSN: 1225-0112 eISSN: 2288-4505

The advantages of this approach include the absence of harmful emissions into the soil and atmosphere when utilizing adhesives, as well as the potential for multiple reuses when opting for tackifiers. This dual usability, combined with their edibility and eco-friendliness, holds significant promise for enhancing our daily lives.

2. Experiment

2.1. Materials

Basil seeds from India were purchased by DG FARM. DI water (distilled water) and glycerol were purchased by DUKSAN science.

2.2. Preparation of BSM powder

Basil seeds (16 g) are added to 1 L of distilled water with a pH of 7 (water-to-seed ratio of 62.5:1). The mixture is stirred for 2 hours at 60 °C and 750 rpm. The mixture is filtered to separate the swollen basil seeds from the water. The swollen basil seeds are transferred to a mesh-like filter and subjected to centrifugation at 15000 rpm for 1 hour at 25 °C. The separated basil seed mucilage is transferred to a plate. After approximately 24 hours of drying at 50 °C in an oven, it is ground into a powdered form using a mortar and pestle.

3. Results and discussion

Despite numerous advancements, the potential of basil seed-based adhesives remains insufficiently explored. Adhesives undergo a transformation from a liquid to a solid state, exhibiting strong adhesive properties that result in irreversible bonding after complete drying. Also, a tackifier immediately establishes bond strength upon contact under low-pressure conditions and maintains reusability even after detachment. BSM demonstrates versatile applicability, serving as a suitable candidate for both adhesive and tackifier applications. When immersed in water, basil seeds swell into gelatinous masses with high water-absorption capabilities. The objective of this study is to extract mucilage from basil seeds (BS) and develop a process for employing BSM as an adhesive material, thus exploring its potential for innovative, edible, and environmentally friendly adhesives based on BSM (Figure 1). The investigation also encompasses the assessment of the physical and mechanical characteristics of adhesives utilizing water and glycerol solvents. While it exhibits the drawback of low water resistance due to the use of water as a solvent, this limitation implies the potential for reusability through rehydration, presenting opportunities for further applications.

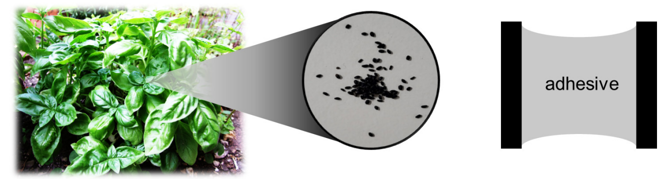

Figure 1. Illustration of the use of basil seeds as adhesive.

The extraction process of BSM is described in Figure 2a. The Basil seeds (16 g) are added to 1 L of distilled water with a pH of 7 (water-to-seed ratio of 62.5:1). The mixture is stirred for 2 hours at 60 °C and 750 rpm. The mixture is filtered to separate the swollen basil seeds from the water. The swollen basil seeds are transferred to a mesh-like filter and subjected to centrifugation at 15000 rpm for 1 hour at 25 °C. The separated basil seed mucilage is transferred to a plate. After approximately 24 hours of drying at 50 °C in an oven, it is ground into a powdered form using a mortar and pestle[5]. We determined the dried BSM yield derived from basil seeds using the method described above. Our findings revealed that roughly 0.05 g of dried BSM powder can be obtained from every 1 g of basil seeds. When this

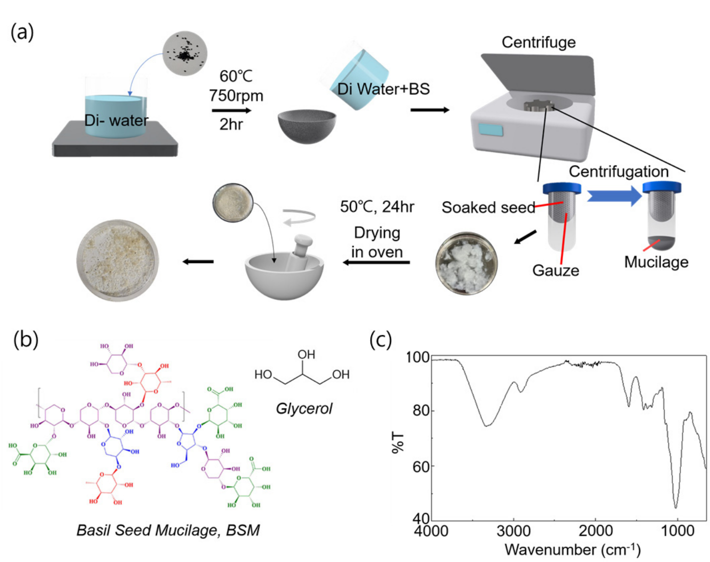

Figure 2. a) Extraction process of BSM using centrifugation method. b) Chemical structure of raw materials used to manufacture basil seed mucilage (BSM) adhesives. c) FT-IR spectra of BSM.

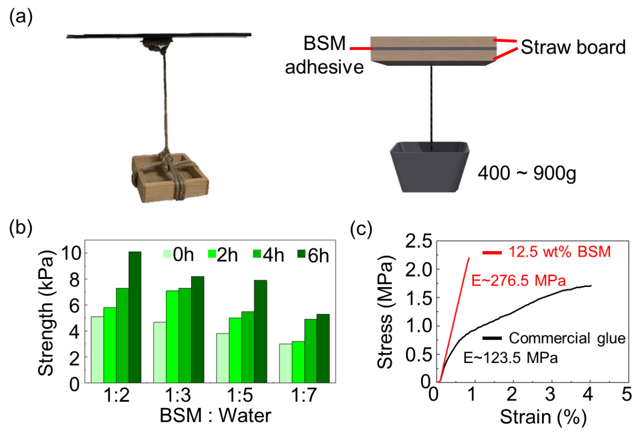

Figure 3. Adhesive strength and mechanical properties according to the evaporation of solvent. a) Actual experimental sample image and schematic representation of how to measure the adhesive strength. b) Bar graph of adhesive strength according to the ratio of BSM to water. The BSM powder was mixed with water at ratios of 1:2, 1:3, 1:5, and 1:7 (BSM powder:water), respectively. As the solvent evaporated and the adhesive solidified over time, adhesive strength increased. c) Stress-strain curves of the 12.5wt% BSM and commercial glue stick with tensile. The Young's modulus of 12.5wt% BSM determined at 0.2% strain was 276.5 MPa. The Young's modulus of commercial glue stick determined at 0.2% strain was 123.5 MPa.

basil seed powder is combined with water at a ratio of 1:5, it can produce approximately 30 g of adhesive per 100 g of basil seeds. This indicates that a significant amount of adhesive can be produced compared to commercial adhesives at a competitive cost. To produce 100g of BSM adhesive, 333 g of seeds are required, costing 3,320 KRW. This cost excludes expenses incurred for solvents and manufacturing processes. Considering that the commercial adhesives cost approximately 4,000 KRW per 100g, a systematic production process can ensure sufficient cost competitiveness.

In the BSM spectrum (Figure 2c), the peaks at 3420, 2923, 1610, 1420 and 1058 cm^{-1} are associated with OH bands of carboxylic acid, —CH_2— and —CH— stretching and bending vibrations, C=C stretching vibration, C=N stretching vibrations, and C–O–C stretching vibrations of glycosides, respectively.[2,4]

In everyday life, considering the most common substrate, paper, as the adherend, experiments were designed (Figure 3a). Strawboard (3 × 3 cm^2) was used as the adherend to measure adhesive strength. The experiments were conducted by suspending baskets on adhesive-coated substrates, with a uniform application of 0.35 g of adhesive for all samples. The experiments involved incrementally adding weight to the baskets at specific time intervals until the substrates detached. The minimum strength observed was approximately 320 g at 0 hours for a 1:7 (BSM:water) ratio, while the maximum strength of approximately 920 g was achieved at 6 hours for a 1:2 (BSM:water) ratio.

The previously obtained BSM powder was mixed with water at ratios of 1:2, 1:3, 1:5, and 1:7 (BSM powder:water) to create adhesive samples. These samples were then applied in a quantity of 0.35 g between the adherends. Over time, as the solvent (water) evaporated,

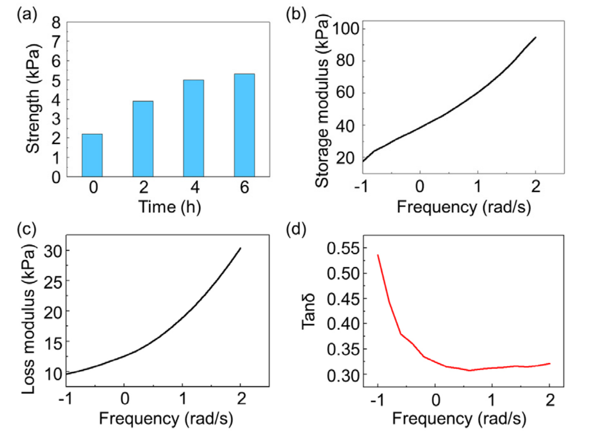

Figure 4. a) Bar graph of adhesive strength of BSM with glycerol according to the time (A ratio of 2:2:1 for glycerol:water:BSM powder). b-d) Rheology tests of a ratio of 2:2:1 for glycerol:water:BSM powder (b: storage modulus, c: loss modulus, d: tan δ).

changes in adhesive strength due to evaporation were measured. As shown in the Figure 3a, the method involved measuring the weight at which the two adherends detached when suspended by a thread at specific time points. As the solvent evaporated and the adhesive solidified over time, adhesive strength increased, with some cases showing more than a twofold difference in strength (Figure 3b).

When comparing the BSM adhesive (with a BSM powder:water ratio of 1:7) to commonly used adhesives in everyday life on a wooden substrate, we conducted tests using a Universal Testing Machine (UTM) in Figure 3c. This BSM adhesive, considered safe for humans and environmentally friendly, demonstrated sufficient strength when evaluated for practical adhesive applications. However, caution is required in extremely dry environments, such as an oven (conditions characterized by temperatures exceeding 50 degrees Celsius, excellent ventilation, and extreme aridity), where solvents(water) completely evaporate, leaving only the matrix (BSM Powder), resulting in a reduction of adhesive strength[6].

Glycerol was introduced to enable the utilization of the adhesive as a tackifier rather than for one-time-use. In contrast to adhesives that cannot be reattached once separated following solidification, glycerol was incorporated to function as a tackifier. Glycerol has been reported to have no adverse effects, and due to its capacity to attract moisture from the environment and inhibit moisture evaporation, it was utilized to suppress solvent evaporation and maintain adhesive strength[7,8]. Similar to the schematic in the Figure 3a, an experiment was conducted to measure adhesive strength over time. Within the first 4 hours, there was a trend of increasing adhesive strength due to minor solvent evaporation (Figure 4a). However, the variation was not significantly different from when using water as the sole solvent. Between 4 to 6 hours, adhesive strength remained relatively stable. With the application of optimal ratios and other additives, the consideration of adhesive utilization as a tackifier becomes a viable option.

At 25 °C, an adhesive with a ratio of 2:2:1 for glycerol:water:BSM

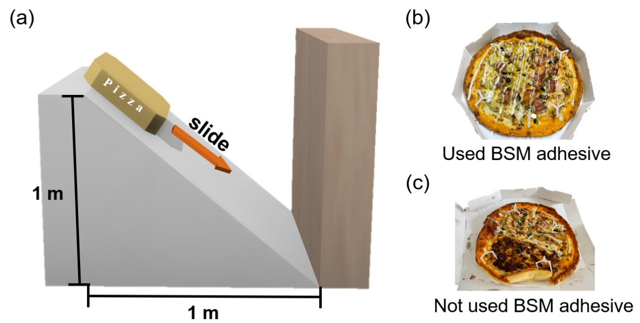

Figure 5. Applying BSM adhesive between the paper at the bottom of a pizza and the pizza box to prevent shifting during delivery. a) Schematic illustration of sliding experiment of the pizza boxes. c) The pizza box with BSM adhesive. d) The pizza box without BSM adhesive.

powder was prepared, and its rheological behavior was assessed using the ARES-G2 and Discovery Hybrid Rheometer (Figure 4b-d). The rheology tests were conducted to directly characterize the response of the pressure-sensitive adhesive (PSA) at various deformation times related to shear resistance, adhesion strength, and peel behavior. The ability to conform to the substrate decreases as the modulus becomes too large. The low G' (storage modulus) and high tan δ values concerning frequency indicate that the adhesive is more easily deformable upon contact with the substrate. For improved adhesion, a higher tan δ is necessary, signifying that G" (loss modulus) is larger than G', indicating that the polymer releases energy through its inherent deformation. This allows the material to establish a strong contact with the substrate, facilitating effective adhesion.[9,10]

Numerous advancements have been investigated in the field of edible adhesives, but BSM provides an eco-friendly alternative with a straightforward extraction process utilizing water as a solvent. This approach allows users to directly control concentration and potentially facilitates recycling. While a higher solvent content tends to weaken adhesive strength (refer to the Figure 3b), the formulation is notably softer, enabling more precise and localized application.

In line with the objective of creating adhesives suitable for individuals prone to allergic reactions and safe for use in food, considerations were made for practical applications in daily life, leading to conducted experiments. Unless an individual exhibits specific allergic reaction to basil, it can be used for potential applications such as non-toxic stickers, toy adhesives for children who can handle items in their mouths without concern, and even for toddler-friendly craft adhesives. To demonstrate its practical utility in everyday life, we conducted experiments. Among two pizza boxes, one remained untouched, while on the other, BSM adhesive was applied at four points along the circumference, 1 g each, between the pizza base paper and the box. Subsequently, to simulate rough delivery conditions, we vigorously shook each pizza box from side to side by 0.5 meters, repeating this motion 10 times and slid them down a 1m by 1m incline (Figure 5a). As seen in Figure 5b, using the adhesive allows the pizza box to remain stable and maintain the shape of the pizza without distortion from shaking or impact. In contrast, as shown in Figure 5c, without adhesive, noticeable shape deformation occurs due to the shaking, an experience familiar in our daily lives when ordering pizza. Thus, BSM adhesive provides an environmentally friendly and harmless way to preserve the shape of pizzas during delivery. This application eliminates the need for pizza securing pins. BSM adhesive not only serves as an eco-friendly adhesive but also offers a solution to the global issue of plastic waste.

4. Conclusion

Current research has indicated the potential utilization of basil seed mucilage as an adhesive that remains stable for both food and natural environments. Basil mucilage presents an intriguing component for the development of innovative adhesives. Basil seeds offered a substantial 20.5% (w/w) content of mucilage, making them highly suitable for use with both water and glycerol solvents due to their hydrophilic nature. The mucilage extracted from basil seeds was dried and processed into the powdered form, and experiments demonstrated its adhesive capabilities by capitalizing on its exceptional water-absorbing capacity. The BSM adhesives, with only a small quantity of 0.35 g, demonstrated the capability to withstand forces approaching 1 kg, indicating that they have adhesive properties exceeding those of commercial glue in everyday life. This study investigated the variation in adhesive strength concerning the moisture content, the solvent, of the BSM adhesive. The findings validated that adhesive strength increases as the solvent evaporates, aligning with the adhesive's inherent characteristics. Furthermore, the introduction of glycerol was explored to sustain adhesive strength, suggesting the adhesive's potential as a tackifier. The study also was conducted involving the secure attachment of pizza boxes and support paper using BSM adhesive, followed by subjecting them to impact tests, thereby assessing their practical applicability for everyday use. This investigation underscores the eco-friendly advantages of BSM adhesive and highlights its potential as an edible adhesive with promising prospects.

Acknowledgement

This work was supported by the Global Joint Research Program funded by the Pukyong National University (202411710001).

References

1. N. Khazaei, M. Esmaiili, Z. E. Djomeh, M. Ghasemlou, and M. Jouki, Characterization of new biodegradable edible film made from basil seed (*Ocimum basilicum* L.) gum, *Carbohydr. Polym.*, **102**, 199-206 (2014).
2. M. Komijani, M. Mohebbi, and B. Ghorani, Assembly of electrospun tri-layered nanofibrous structure of zein/basil seed gum/zein for increasing the bioaccessibility of lycopene, *LWT*, **161**, 113328 (2022).
3. S. Nazir and I. A. Wani, Functional characterization of basil (Ocimum basilicum L.) seed mucilage, *Bioact. Carbohydr. Diet.*

Fibre, **25**, 100261 (2021).
4. S. A. Ghumman, S. Noreen, H. Hameed, M. A. Elsherif, R. Shabbir, M. Rana, K. Junaid, and S. N. A. Bukhari, Synthesis of pH-sensitive cross-linked basil seed gum/acrylic acid hydrogels by free radical copolymerization technique for sustained delivery of captopril, *Gels*, **8**, 291 (2022).
5. S. Nazir, I. A. Wani, and F. A. Masoodi, Extraction optimization of mucilage from Basil (*Ocimum basilicum* L.) seeds using response surface methodology, *J. Adv. Res.*, **8**, 235-244 (2017).
6. S-Y. Choe, 접착의 원리와 이론, *Elastomers and Composites*, **25**, 291-309 (1990).
7. K. Takamura, H. Fischer, and N. R. Morrow, Physical properties of aqueous glycerol solutions, *J. Pet. Sci. Eng.*, **98**, 50-60 (2012).
8. X. Lin, W. Ma, L. Chen, L. Huang, H. Wu, and A. Takahara, Influence of water evaporation/absorption on the stability of glycerol–water marbles, *RSC Adv.*, **9**, 34465-34471 (2019).
9. Mazzeo, F. A., Characterization of pressure sensitive adhesives by rheology, *TA Instruments Report RH082*, **1-8** (2002).
10. Rafe, A. and Razavi, S. M., Dynamic viscoelastic study on the gelation of basil seed gum, *Int. J. Food Sci. Technol.*, **48**, 556-563 (2013).

Authors

Hyun Yun Lee; B.Sc., Researcher, Department of Polymer Engineering, Pukyong National University, Busan 48513, Korea; angyun12@naver.com

Hye Joo Lee; B.Sc., Master Student, Department of Polymer Engineering, Pukyong National University, Busan 48513, Korea; hjl7155@naver.com

Seong Ho Ryu; Ph.D., Senior Researcher, Applied Polymer Research Center, Korea Conformity Laboratories, Incheon 21594, Korea; ryush@kcl.re.kr

Daniel Sim; Ph.D., Senior Scientist, Integrative Health & Performance Sciences Division, UES, Inc, Dayton, OH, 45432 USA; jksimtt@gmail.com

Dae Seok Kim; Ph.D., Associate Professor, Department of Polymer Engineering, Pukyong National University, Busan 48513, Korea; daeseok@pknu.ac.kr