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From waste to tape: Inverse vulcanization of sulfur and solvent-based depolymerization for preparation of pressure-sensitive adhesives



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ABSTRACT

Upcycling is an eco-friendly method that turns waste into valuable items. Elemental sulfur, a waste substance produced in large amounts (70 million tons per year), can be converted into valuable polymers using inverse vulcanization. However, the traditional method had significant limitations, such as limited consumption and weak strength. To address this problem, we newly fabricated sulfur-based pressure-sensitive adhesives (PSAs). PSAs are soft and tacky materials that should have low modulus, and they are used extensively (3.1 million tons per year). We prepared sulfur-based PSAs in four steps: polymerization, dissolution, coating/drying, and curing. In the polymerization step, castor oil and oleic acid were employed to facilitate inverse vulcanization. The resulting polymer then underwent solvent-based depolymerization to be coated in a film form. After drying and ultra-violet (UV)-induced curing, the tapes were successfully produced. Significantly, we adjusted the PSA's crosslinking density by changing the oleic acid content and UV dose. As the amount of mono-functional oleic acid decreased or the UV dose increased, the resulting PSA's crosslinking density increased. The best conditions led to PSAs with excellent adhesion performances and creep resistance, similar to commercial tapes. Moreover, the prepared PSAs and conductive sensors.

1. Introduction

A distinctive technique called upcycling holds the potential to transform waste into valuable products, offering a promising avenue toward achieving environmental sustainability. Elemental sulfur, with an annual production of 70 million tons resulting from the desulfurization of crude oil and natural gas [1], represents one such waste stream that warrants consideration. Despite its direct utilization in fertilizers and the rubber industry, substantial quantities of surplus sulfur accumulate annually due to its limited consumption.

The domain of polymers holds the most auspicious potential for the large-scale upcycling of sulfur waste [2]. The most straightforward technique to create sulfur polymers involves self-polymerization initiated by thiyl radicals, which emerge through the homolytic cleavage of

S–S bonds. Nevertheless, the utilization of this method to generate sulfur-based polymers faces challenges due to the depolymerization caused by the backbiting of terminal sulfur radicals [3]. To address this issue, Pyun and coworkers [4] introduced the concept of inverse vulcanization, employing multi-functional crosslinkers to curb the undesirable recombination of terminal sulfur radicals. Since the inception of this groundbreaking approach to inverse vulcanization, extensive research has been undertaken to produce high-value products endowed with distinctive functionalities like electrochemical properties [4–7], dynamic covalent bonding [8–10], infrared transmittance [10–14], metal sorption [15–20], and adhesive property [21–23]. However, these applications typically necessitate only marginal quantities in global production, falling short of adequately consuming the surplus sulfur. A notable limitation of most polymers synthesized via inverse

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Abbreviations: PSA, Pressure-Sensitive Adhesive; T_g, Glass transition temperature; THF, Tetrahydrofuran; UV, Ultraviolet; DSC, Differential scanning calorimetry; G', Stoorage modulus.

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Fig. 1. a) Schematic illustration for preparing tape from waste via inverse vulcanization of sulfur and solvent-based depolymerization. b) Chemical structure of the starting materials and prepared tape.

vulcanization is their deficient mechanical strength, significantly constraining their potential applications.

Pressure-sensitive adhesive (PSA) is a polymer-based material that exhibits excellent adhesion by light pressure owing to its inherent tackiness [24,25]. This can be considered the best candidate for the potential application of sulfur-based polymer for two reasons. Firstly, most PSAs exhibit lower mechanical strength than other polymer-based materials. This is because the storage modulus of PSA necessitates appropriate reduction to facilitate optimal interfacial wetting between the PSA and the substrate [26–28]. Secondly, the substantial global production scale of PSAs (amounting to 3.1 million tons in 2016) [29] further supports their utility.

Commonly, PSAs are supplied as "tape", comprising a backing film coupled with a PSA layer. Preparing a PSA layer on a backing film requires the application of a polymer resin in a liquid state, followed by its solidification. In this procedure, incorporating organic substances, water, or prepolymer is feasible to render the PSA resin amenable to coating [25]. Notably, the viscous prepolymer can be prepared through methods such as inverse vulcanization utilizing monoene [8] or step curing [30], yet controlling the viscosity of the prepolymer resin and the degree of crosslinking in the final products presents challenges.

Herein, we introduce a novel approach involving inverse vulcanization and solvent-based depolymerization for the purpose of upcycling sulfur waste into PSAs. The synthetic procedure, shown in Fig. 1a, comprises four distinct steps: (i) polymerization, (ii) dissolution, (iii) coating/drying, and (iv) curing. In the polymerization step, inverse vulcanization was conducted using castor oil and oleic acid (Fig. 1b). The resultant polymer was dissolved within a solvent to produce PSA in a film form. Next, the crosslinking degree, reduced for dissolution, was increased again through ultra-violet (UV) curing after film drying. This comprehensive methodology facilitates rapid, thin, and uniform film production, and the resulting PSAs exhibit sufficient adhesion to be used as tape. Additionally, dynamic covalent bonds imparted self-healing properties to the PSAs, further extending their potential utility.

2. Results and discussion

2.1. Design strategy

Two primary considerations are essential for the production of sulfur-based tape formulation. First, it is crucial to utilize reactive compounds containing multiple vinyl groups to induce the polymerization of elemental sulfur (\geq 99.0 %, Sigma-Aldrich) through inverse vulcanization. Castor oil (Sigma-Aldrich), an inexpensive and sustainable resource, was employed to facilitate inverse vulcanization [31–35]. Second, given that PSAs exhibit adhesive properties at typical application temperatures, careful attention must be given to their glass transition temperature (T_g), ideally 25–45 °C lower than the usage temperature [36]. To address this requirement, oleic acid (Sigma-Aldrich), featuring one vinyl group in a single molecule, was employed to adjust the resulting polymer's T_g. These two monomers possess notably high boiling points (313 °C for castor oil and 360 °C for oleic acid), thereby enabling inverse vulcanization, executed at an elevated temperature of 170 °C.

Following polymerization, the resultant polymer was dissolved in tetrahydrofuran (THF), formulating a viscous resin through the cleavage of disulfide bonds within the chain structure. The derived resin was subsequently applied in film form onto a 50 μ m-thick polyethylene terephthalate backing film. After that, the solvent and H₂S gas were eliminated during the drying step (100 °C, 2 min). However, due to the relatively low extent of crosslinking exhibited by the PSAs post the dissolution and drying process, their capacity to endure high temperatures and creep were diminished, making them unsuitable for use as commercial tape [37–41]. Consequently, the fractured disulfide bonds underwent a re-crosslinking process to enhance the thermal and creep resistance of the resultant PSA layers. This crosslinking step was driven by UV light instead of heat to prevent any possible damage to the backing film due to high temperatures.

2.2. Polymerization and dissolution behavior

For inverse vulcanization, elemental sulfur was mixed with castor oil



Fig. 2. a) Inverse vulcanization behavior (solidification and conversion) and b) properties of the synthesized polymer (T_g and gel content). c) Change of gel content as a function of dissolution time. The elemental sulfur content was fixed at 25 %.

and oleic acid. The proportion of elemental sulfur by weight remained constant at 25 %, while two different monomers were employed in varying ratios. The inverse vulcanization was conducted at 170 °C with vigorous stirring. After obtaining a rubber-like product (Fig. S1), the reaction was terminated by cooling to room temperature (RT = 25 °C). Incorporating multi-functional castor oil resulted in a crosslinked network, whereas the mono-functional oleic acid led to a linear polymer chain. Consequently, as the quantity of oleic acid was augmented, the solidification time was prolonged, extending from 46 to 76 min (Fig. 2a). To assess the extent of inverse vulcanization, the reduction in the number of C=C bonds within the monomers was estimated through nuclear magnetic resonance spectroscopy (Fig. 2a and Fig. S2). As the amount of oleic acid was increased, the final conversion experienced a slight rise, elevating from 54.3 to 82.9 %. This phenomenon was attributed to the extended reaction time stemming from the presence of oleic acid.

Next, the synthesized polymer's glass transition temperature (T_g) was evaluated using differential scanning calorimetry (DSC). As the oleic acid led to the formation of a linear polymer structure rather than a crosslinked network, the T_g displayed a reduction with increasing oleic acid content (Fig. 2b). Once the proportion of oleic acid surpassed 10 wt %, the resultant polymers demonstrated a notably diminished T_g value (–27.7 °C), making them suitable for application as PSAs under RT

conditions. Moreover, the DSC results validated the degradation stability, as evidenced by the absence of a distinct melting peak corresponding to elemental sulfur after 24 h of storage (Fig. S3).

After the inverse vulcanization, the resultant polymer was dissolved in THF at a fixed concentration (10 %) and temperature (50 °C). Immediately after dissolution, the gel content decreased with oleic acid augmentation, but all showed an 80 % or more gel content (Fig. 2b). As time elapsed, the gel was gradually decomposed (Fig. S4), accompanied by the transformation of the initially opaque solution into a transparent state (Fig. S5). The synthesized polymer without oleic acid maintained a high gel content (>80 %) even after 72 h (Fig. 2c). However, upon exceeding a 10 wt% proportion of oleic acid, the resulting polymer was fully dissolved within 48 h. This result elucidates that incorporating oleic acid expedited the dissolution process, reducing overall process time.

2.3. Rheological properties and adhesion performances

In the inverse vulcanization and dissolution, it was established that introducing oleic acid at concentrations of 10 wt% or higher led to the polymer produced exhibiting sufficiently reduced T_g and enhanced dissolution characteristics. Consequently, PSAs were formulated incorporating 15 and 20 wt% of oleic acid, and their rheological properties



Fig. 3. Temperature sweep test results of the cured PSAs containing a) 15 wt% and b) 20 wt% of oleic acid. Creep and recovery test results of the cured PSAs containing c) 15 wt% and d) 20 wt% of oleic acid. e) Appearance of the prepared PSAs, f) test scheme, and g) results for the 180° peel test. Detailed information is described in supporting information.

and adhesion performances were subjected to analysis. The dissolved polymer solution was applied as a film with a thickness of 25 μ m, followed by a drying step at 100 °C lasting 2 min. Subsequently, the dried PSA films underwent curing via exposure to UV radiation (intensity: 500 mW/cm²) to regenerate a crosslinked network structure via UV-induced dynamic exchange of S–S bonds [42–44].

The initial step involved conducting a temperature sweep test to confirm the PSAs' Tg and high-temperature modulus (Fig. 3a and b). When the amount of UV dose increased from 0 to 5 J/cm^2 , there was a corresponding increase in the stiffness of the crosslinked structure, as indicated by the plateau modulus. This result suggests that UV light effectively induced the formation of the crosslinked network, enhancing the high-temperature modulus and crosslinking density (Fig. S6). Comparing cured PSAs containing 15 and 20 wt% of oleic acid, those with 15 wt% exhibited a higher plateau modulus and crosslinking density. This difference can be attributed to the property of oleic acid to induce the formation of a linear polymer structure. Concerning the T_o, no notable variations were observed among the cured PSAs. While PSAs with 15 wt% of oleic acid displayed a slightly elevated T_g (-18.5 °C) compared to those with 20 wt% of oleic acid (-20.2 $^{\circ}$ C), the difference was minimal. Furthermore, exposure to UV light did not substantially alter the Tg and all PSAs exhibited sufficiently low Tg values, making them suitable for application as commercial tape.

The verification of a crosslinked structure resulting from UV exposure was also corroborated through the "creep and recovery" test results (Fig. 3c and d). As the amount of UV dose increased, creep deformation was strictly suppressed, and the strain was rapidly recovered after removing the applied stress. Additionally, the adhesive formulated with 15 wt% of oleic acid displayed reduced creep deformation and superior recovery compared to the 20 wt% variant. This improvement can be attributed to the higher crosslinking density in the adhesive with 15 wt% of oleic acid. In other words, the system we established effectively controls the degree of crosslinking density by adjusting the oleic acid content and the UV dose. Importantly, our method could be useful for making adhesives for flexible displays where precise control of the adhesive's crosslinking density is crucial [40,41].

After assessing the rheological properties of the PSAs, we fabricated the PSAs into a tape structure that consisted of a backing film bonded with a PSA layer. The prepared tapes were slightly yellowish but transparent (Figur 3e). Adhesion performances were evaluated by a 180° peel test (Fig. 3f), employing stainless steel as the substrate. With increased crosslinking density due to exposure to UV irradiation, peel strength rose due to heightened cohesive strength (Fig. 3g). When the oleic acid content was 20 wt%, and the UV dose was 2 J/cm^2 , the resulting PSA exhibited high adhesion strength (peel strength: 2.96 N/ cm) and creep resistance (creep strain: 13.3 %). Notably, this peel strength value closely resembles that of tapes available in the market [45-47], suggesting the potential for our approach to yield commercially viable tapes. However, subjecting the tapes to excessive UV irradiation (5 J/cm²) significantly reduced the peel strength due to diminished surface wetting. The decline was more pronounced in the tapes containing 15 wt% oleic acid than those with 20 wt%, owing to the higher level of crosslinking density [37]. This result underscores the critical importance of optimizing UV exposure conditions and adhesive formulation to attain optimal adhesive performance.

Self-healing property of the prepared PSAs

a) Oleic acid: 15 wt%



b) Oleic acid: 20 wt%



Fig. 4. The prepared PSAs' self-healing properties contain a) 15 wt% and b) 20 wt% of oleic acid. Detailed information is described in supporting information.

2.4. Self-healing property

As the S–S bonds in the polymer can be easily exchanged by heat, the PSAs we prepared were expected to exhibit self-healing properties. A rheology test was conducted to assess the self-healing properties of the prepared PSAs. This test involved two main steps: a damaging step (applied strain and time: 500 % and 100 s) and a recovery step (applied strain and time: 1 % and 300 s). During the damaging step, there was a sharp decline in the storage modulus (G'), indicating that the internal bonds of the PSA broke due to the high strain. Subsequently, with lower strain, the G' value increased again during the recovery step, suggesting that the PSAs had healed themselves. As shown in Fig. 4, the ability of the PSAs to self-heal was strongly related to the healing temperature rather than the oleic acid content. At a low temperature (60 °C), the PSAs with 15 and 20 wt% of oleic acid were not sufficiently healed, showing a decrease in G' of the third recovery step. Remarkably, at a temperature of 100 °C, the PSAs exhibited exceptional self-healing characteristics, displaying a higher G' value after healing than the original value before the damaging step. This outcome can be attributed to additional bonding formed by the unreacted C=C in oleic acid and castor oil, effectively creating more crosslinks in the PSA. Through our approach, PSAs capable of self-healing were successfully prepared. We believe this methodology can fabricate functional PSAs, including remoldable PSA [48] and conductive sensors [49,50].

3. Summary and conclusions

This study presented a comprehensive design strategy for fabricating tapes from industrial waste elemental sulfur. Preparation of pressuresensitive adhesives (PSAs) from elemental sulfur consisted of four steps: (i) polymerization, (ii) dissolution, (iii) coating/drying, and (iv) curing. In the polymerization step, castor oil and oleic acid were employed to facilitate inverse vulcanization. The resulting polymer was then dissolved in tetrahydrofuran, and the solution was coated onto a backing film. Afterward, the solvent was dried, and the prepared film was exposed to ultra-violet (UV) light to regenerate crosslinks that enhance thermal and creep resistance. Crosslinking density was adjustable by changing the amount of oleic acid and UV dose, which influenced the rheological properties and adhesion performances of the PSAs. Notably, the optimized conditions (20 wt% oleic acid, 2 J/cm² UV dose) vielded PSAs with excellent adhesion performances and creep resistance. This finding demonstrates the potential of our method to produce PSAs suitable for commercial tapes. Furthermore, these PSAs exhibited self-healing properties due to dynamic covalent bonds (S-S), suggesting the feasibility of innovative applications like remoldable PSAs and conductive sensors.

CRediT authorship contribution statement

Seong-Ju Lee: Conceptualization, Investigation, Writing – original draft. **Gi-Yeon Han:** Methodology. **Mo-Beom Yi:** Methodology. **Jong-Ho Back:** Writing – review & editing, Funding acquisition. **Hyun-Joong Kim:** Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jmrt.2024.01.218.

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