Proceedings of the 2nd International Conference on Functional Materials and Civil Engineering
DOI: 10.54254/2755-2721/91/20241091

Study on UV-Curable Acrylic Clear Viscoelastic Materials with Grafted Structures

Junqi Liu

School of Advanced Soft Matter, South China University of Technology, Tianhe District, South China University of Technology, Duxing Building (Building No. 2) North Tower 102, Guangzhou, China

junqilius@outlook.com

Abstract. Flexible substrates are crucial for wearable electronic strain sensors, but achieving biocompatibility, low cost, and aesthetic appeal alongside good mechanical performance remains challenging. Acrylic-based clear viscoelastic films (CVFs) are gaining attention due to their excellent transparency and potential for enhanced mechanical properties. This study synthesized copolymers CVF1-4 with varying branch content using soft monomers 2-ethylhexyl acrylate (2-EHA), butyl acrylate (n-BA), and functional monomer acrylic acid (AA). Employing a "grafting-through" strategy, small molecule monomers, ATRP-synthesized oligomer pBA25, and photoinitiator 1,6-hexanediol diacrylate (HDDA) were in-situ polymerized under UV light to form transparent, flexible elastic films CVF1-4. Between -20 to 80 °C, CVF1-4 demonstrated excellent mechanical properties such as low modulus (in the kPa range), low glass transition temperature (below -20 °C), large deformation (500-600%), and high strain recovery rate (>95%). Among them, CVF3 with 15% branching showed a balanced comprehensive performance, exhibiting primarily elastic behavior at room temperature. Therefore, selecting appropriate molecular composition and grafting structure provides a simple and customizable solution for optimizing CVF properties.

Keywords: Clear viscoelastic materials, Grafting, Flexibility, Photocrosslinking.

1. Introduction

Wearable electronic devices are cutting-edge products in artificial intelligence for health monitoring, diagnosis, and portable electronics [1]. As core components, electronic sensors capture physiological signals and convert them into visual electronic signals, effectively detecting human activities. Ideal wearable electronics need excellent softness to adapt to muscle movements and avoid irreversible deformations like bending, wrinkling, or breaking during use. Since they are in direct contact with the skin, they must also have low biotoxicity and removability, sometimes requiring high transparency for aesthetic reasons. However, traditional electronic sensors based on metallic conductors and other inorganic materials struggle to combine these advantages.

Organic materials for flexible strain sensors are typically lightweight and aesthetically pleasing with adjustable mechanical properties. Thermosetting resins like polyolefin elastomers are widely used in the industry due to their low cost, aging resistance, and stability. Solvent-based production methods may include plasticizers, potentially releasing harmful substances like formaldehyde. To balance softness

DOI: 10.54254/2755-2721/91/20241091

and toughness, achieving reversible large deformations within the normal use temperature range requires attention to the composition and structure of the polymers.

2. Research Status and Background

The clear viscoelastic film (CVF) is a flexible polymer with excellent optical properties, such as high light transmittance and low haze, without the need for additional additives. It has been applied as an adhesive for foldable smartphone screens [2]. CVF is essentially a viscoelastic material, where there is always a trade-off between elastic and viscous properties. For instance, increasing the strain recovery rate often negatively impacts the elongation at break. To enhance reversible recovery and various mechanical strengths, the main approach currently is to incorporate additional processing agents and plasticizers into the CVF network to improve cohesion. However, many additives are cytotoxic and may reduce transparency, thereby limiting the material's application [3].

Grafting methods can improve or customize polymer properties without using additives or altering chemical composition. This is achieved by chemically bonding macromolecular branches or functional side groups to the polymer backbone, producing products that exhibit characteristics of both the main chain and side chains [4]. By properly controlling the branch composition, various properties such as strength and ductility can be effectively improved. This effect is similar to adding plasticizers, but graft structures do not migrate or leach out over time, preventing surface contamination and supporting the requirements of "green chemistry" [5].

The monomer composition, including types and content, affects various properties of acrylic CVF. For example, Seung-Suk Baek et al. [6] found that the structure of acrylic monomers influences the rigidity of the polymer backbone. The larger the free volume of substituents such as side chains, the lower the cohesion of the polymer, resulting in a decrease in tensile strength and other elastic properties. Ju Hak Lee et al. [7] used UV curing to copolymerize EHA with MA, AA, and HEA, respectively, and found that polymers containing AA had higher peel strength; adding MA achieved instant strain recovery with nearly zero hysteresis loss; however, organic solvent ethanol negatively affected the mechanical and optical properties of the copolymers.

Molecular weight is also associated with the viscoelasticity of acrylic CVF. Jung-Hun Lee et al. [8] found that increasing molecular weight can enhance intermolecular interactions through chain entanglement, as evidenced by various thermally initiated acrylic copolymers with molecular weights ranging from 86 kDa to 108 kDa, showing significantly enhanced viscoelasticity. Raghunath P. Ingale et al. [9] used reversible addition–fragmentation chain transfer polymerization (RAFT) to obtain EHA emulsion-type polymers with molecular weights of 2×105, 7×105, 14×105, and 20×105 Da. Their shear strength positively correlated with molecular weight, while peel strength first increased and then decreased, with the highest peel strength observed in products with a molecular weight of 7×105 Da.

Crosslinking forms network structures between molecular chains, allowing a wide range of adjustments to material properties, making it possible to prepare CVF with varying performance from a single raw material. Based on the reaction conditions, crosslinking agents are divided into thermal crosslinking agents like isocyanates and UV crosslinking agents like 1,6-hexanediol diacrylate (HDDA).

More flexible and elastic acrylic graft polymers have been extensively studied in recent years. Andrew Goodwin et al. [10] used the "grafting through" method, first synthesizing structurally defined poly(methyl methacrylate) (PMMA) side chains through living anionic polymerization, and then using RAFT to randomly copolymerize them with n-BA, achieving a product with a fracture elongation of 170% and a strain recovery hysteresis of less than 15%. Anastasia Mpoukouvalas et al. [11] combined "ATRP" and "grafting through" to introduce short pBA side chains into a uniformly crosslinked HEA network, obtaining a structurally defined super soft elastic material with high flexibility and rebound after large deformation, and a storage modulus in the elastic region of less than 5 kPa.

From the preparation system perspective, acrylic CVF is mainly realized through a free radical polymerization mechanism, primarily including solution polymerization and bulk polymerization. Gang Li et al. [18] explored the effects of different processes on solvent-based acrylic CVF, finding that segmented polymerization products had the best overall performance and higher reaction conversion

DOI: 10.54254/2755-2721/91/20241091

rates compared to graft polymerization and one-shot feeding methods. Heeyeon Moon et al. [12] initiated and in-situ crosslinked under thermal, UV, and combined conditions, successfully conducting solvent-free bulk polymerization to directly obtain acrylic copolymer films. They showed that the combination of both methods favored a more systematic adjustment of crosslink density, resulting in products with balanced viscoelasticity and good optical transparency, providing a feasible approach to obtaining environmentally friendly acrylic CVF.

3. Research Design

The rise of wearable devices has driven research on CVF. Currently, most research on acrylic CVF focuses on linear copolymers, with less attention to the impact of grafting. To explore the relationship between branch number and performance, this study constructed a model of acrylic copolymers with short branches and grafting gradients, aiming to improve deformation degree while balancing reversible recovery performance and mechanical strength, thereby expanding the application of CVF in flexible optical materials. To maintain CVF's transparency and biocompatibility, only acrylic monomers were used as copolymer raw materials and crosslinking agents without adding other additives. By using the "grafting-through" strategy and ATRP, macromolecular monomers with controllable polymerization degree (n = 25) were synthesized and polymerized using a solvent-free UV method to reduce cost and environmental pollution. The study compared the differences in rheological properties, reversible recovery performance, and tensile properties of CVFs with different branch contents, analyzing the impact of network structure and branch number on performance, providing a reference for optimizing CVF performance.

3.1. Specific Plan

A series of grafted acrylic CVFs were designed to systematically study their properties. Soft monomers n-butyl acrylate (n-BA, Tg=-56°C) and 2-ethylhexyl acrylate (2-EHA, Tg=-70°C), and hard monomer acrylic acid (AA, Tg=106°C) were mainly selected, adjusting the content of the three to make the copolymer's Tg below room temperature. The functional monomer AA contains highly polar -COOH, which can increase cohesion; thus, AA content does not exceed 10 wt% to maintain softness. To study the effect of branch content on CVF performance, four gradients of 0%, 5%, 15%, and 30% were set, synthesizing CVF1 (linear control) and CVF2/3/4 (graft structure). Short-chain macromolecular monomer (pBA25, polymerization degree 25) was used for "grafting-through" in-situ polymerization to prepare random copolymers. The composition and structure of the products were analyzed using gel permeation chromatography (GPC), Fourier-transform infrared spectroscopy (FTIR), nuclear magnetic resonance hydrogen spectroscopy (1H-NMR), and tested for tensile strength, creep recovery, rheological properties, and glass transition temperature to comprehensively evaluate the viscoelasticity and flexibility of CVF.

3.2. Technical Route

The designed acrylic CVF is based on a free radical polymerization mechanism. The overall preparation route is shown in Figure 1. Firstly, ATRP is used to prepare macromolecular monomers with the required polymerization degree, which are then polymerized with other small monomers in bulk polymerization, initiated and crosslinked under UV conditions to directly prepare films.

3.3. Technical Route

The acrylic CVF designed in this paper is based on the free radical polymerization mechanism. The overall preparation route is shown in Figure 1. First, the macromolecular monomer with the required degree of polymerization is prepared using ATRP. Then, it is bulk polymerized with other small molecule monomers, initiated and cross-linked under UV conditions, and directly formed into a film.

DOI: 10.54254/2755-2721/91/20241091

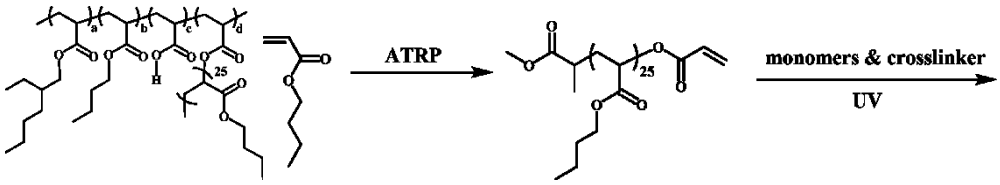
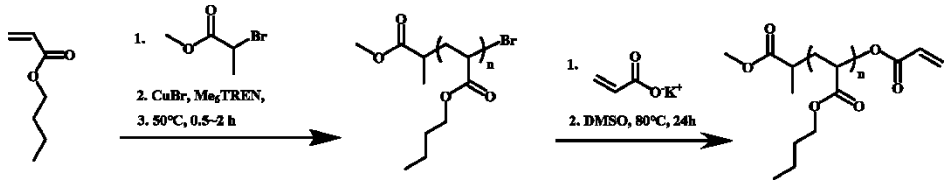


Figure 1. Technical Route

4. Preparation of Transparent Viscoelastic Body

(1)



(2)

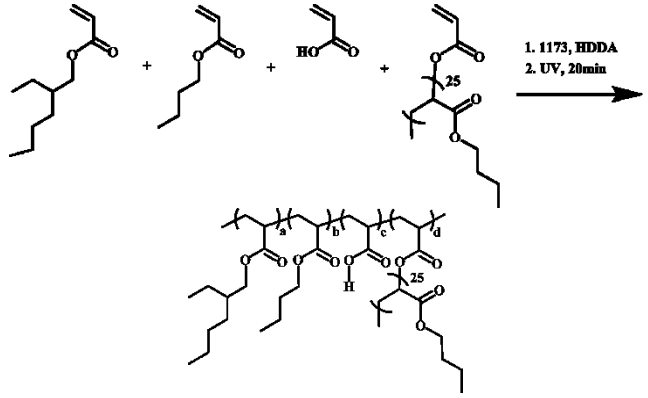


Figure 2. Preparation steps of acrylic viscoelastic body (a) synthesis of macromolecular monomer; (b) synthesis of graft copolymer

Dissolve the catalyst CuBr in purified BA according to the molar ratio [M]0:[I]0:[C]:[L]=25:1:0.05:0.05, add the initiator methyl 2-bromopropionate and Me6TREN, and rapidly inject into an anaerobic reaction system. Heat in a water bath to 50°C, stir at 200 rpm, and react for 20 minutes under nitrogen protection. After the reaction, add EAc to dissolve, filter three times through a sintered glass funnel and basic alumina, filter twice through petroleum ether, and rotary evaporate to obtain pBA25-Br. Dissolve pBA25-Br and potassium acrylate (molar ratio 3:1) in DMSO, stir for 24 hours in an 80°C oil bath for nucleophilic substitution reaction. Pour the product into ice water, stir, and layer, discard the upper aqueous phase, wash with KHCO3 solution to remove DMSO and potassium acrylate. Dissolve in DCM again, break the emulsion with NaCl aqueous solution, extract twice with DCM, dry and filter, and rotary evaporate to finally obtain pBA25.

Weigh the required monomers according to Table 4-3, add 1 g each of photoinitiator 1173 and crosslinker HDDA according to 0.1% of the mass of the acrylic copolymer backbone, and mix the prepolymer. Cut a suitably sized silicone frame and attach it to a 50μm thick silicone release film, clamp with an ultra-white glass plate, and then inject the above mixed liquid into the silicone frame, exhausting the air. Control the total energy of the photoreaction to 3000 mJ/cm², react under UV light with a power of 9.6 mW/cm² for 20 minutes to obtain the formed CVF gel film.

DOI: 10.54254/2755-2721/91/20241091

Table 1. Experimental Formulations

Ingredients	2-EHA(g)	n-BA(g)	AA(g)	pBA25 (g)
CVF_1	4.5	4.5	1	0
CVF_2	4.5	4.5	1	0.5
CVF_3	4.5	4.5	1	1.5
CVF_4	4.5	4.5	1	3

5. Experimental Results Analysis

This paper designs CVF1-4 with a gradient of short side chain mass fractions. The mass ratios of the comonomers are 2-EHA:n-BA:AA = 45:45:10:0, 45:45:10:5, 45:45:10:15, 45:45:10:30. The overall polymerized products are soft, transparent solid films similar in appearance, as shown in Figure 3.

Figure 3. Actual Product Image, from top to bottom: CVF_{1-4}

5.1. Gel Rate Analysis

The gel rate is expressed as the ratio of the remaining mass of CVF1-4 after swelling in a good solvent EA to the initial mass, reflecting the proportion of the cross-linked part. Physical cross-linking is mainly caused by hydrogen bonds between the carboxyl groups of AA and chain entanglement caused by intermolecular interactions; chemical cross-linking is derived from the stable network formed by the reaction with bifunctional HDDA. Table 2 shows that the gel rates of the four samples are all between 93% and 97%, indicating a good degree of cross-linking. Although the pBA25 content is different, due to the bridging effect of HDDA being superior to physical action, the difference in the degree of cross-linking is not significant. The gel rate of CVF4 is the lowest (93.48%), possibly due to a large number of short grafted polymers forming a brush structure, making chain cross-linking difficult. Because highly cross-linked copolymers are difficult to dissolve in solvents suitable for GPC, and the insoluble part exceeds 90%, GPC was not used to measure the molecular weight and distribution of CVF_{1-4}.

Table 2. Gel Rate of Grafted Transparent Viscoelastic Body

Sample	Gel Rate (%)
CVF_1	95.53
CVF_2	94.84
CVF_3	96.83
CVF_4	93.48

DOI: 10.54254/2755-2721/91/20241091

5.2. Structural Characterization

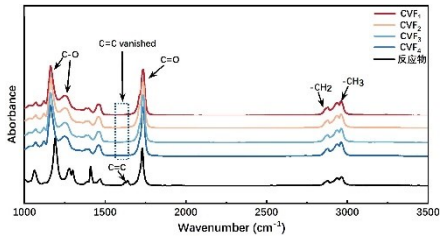


Figure 4. NMR spectra of the macromonomer.

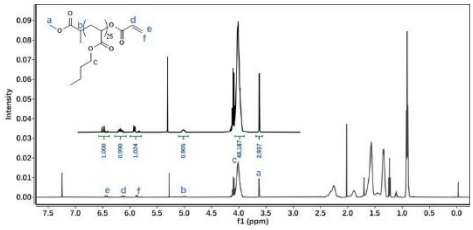


Figure 5. FTIR spectra of the acrylic copolymer and the reactive monomers.

To estimate the length of the grafted side chains, gel permeation chromatography (GPC) was first used to analyze the ATRP product pBA25-Br. The number average molecular weight was found to be 3512 Da, close to the theoretical value of 3385 Da; the weight average molecular weight was 4143 Da, with a polydispersity index of 1.180, indicating minimal chain transfer.

Nuclear magnetic resonance (NMR) spectroscopy revealed that peaks in the 2-2.5 ppm range correspond to the α-H of the carboxyl group in BA, peaks in the 1-2 ppm range are mainly from saturated aliphatic side chains, peaks in the 5.5-6.5 ppm range correspond to C=C double bonds after end group substitution, the peak at 5 ppm corresponds to the α-H of the initiator carboxyl group, the broad peak at 4.1 ppm is attributed to the α-H of the butyl group in BA, and the sharp peak at 3.6 ppm corresponds to -CH3 in the initiator. Combining integration area calculations, the yield of pBA25 was approximately 90.08%, with an absolute degree of polymerization of 24, indicating that the macromonomer was successfully synthesized via an active free radical method with a high yield. Fourier transform infrared spectroscopy (FTIR) analysis showed that the FTIR spectra of CVF1-4 exhibited absorption peaks of -CH3 and -CH2 (2860 cm-1, 2929 cm-1, 2959 cm-1), the carbonyl C=O absorption peak (1731 cm-1), and characteristic peaks of -C-O- (1170 cm-1 and 1250 cm-1). The characteristic peak of the C=C double bond (1636 cm-1) disappeared, indicating that the C=C bond participated in the free radical addition, and the final product no longer contained C=C bonds. In conclusion, the acrylic CVFs were successfully synthesized, conforming to the molecular structure characteristics of the copolymer P(AA-co-2-EHA-co-n-BA).

5.3. Rheological Properties

From the perspective of viscoelasticity analysis, increasing the viscosity of CVF leads to a reduction in elasticity. The viscosity of the polymer is related to the loss modulus (G"), while elasticity is related to the storage modulus (G'). When G" is greater than G', the loss factor (tan δ = G"/G') is greater than 1, indicating that viscosity is dominant, and the polymer will exhibit stress relaxation under external force, leading to energy dissipation and irreversible deformation when the external force is removed, which is unfavorable in practical applications.

Considering the operating temperature range of wearable electronics from -20°C to 80°C, this study tested the rheological behavior of four CVF samples from -50°C to 150°C, obtaining curves of storage modulus (G'), loss modulus (G"), and loss factor (tan δ) with temperature (see Figure 5 and Table 3). Figure 5(a) shows that the storage modulus of the four samples is large at low temperatures, rapidly decreases with increasing temperature, reaches its maximum at -50°C, approaches zero at room temperature, and then remains stable. The variation trend of the loss modulus with temperature is similar in Figure 5(b).

At -50°C, 25°C, and 150°C, the moduli of CVF1-4 decrease sequentially, with CVF1 having the highest modulus and CVF4 being the softest but with poorer mechanical performance. As the graft content increases, the polymer chains' mobility enhances, making chain relaxation or slippage easier. The moduli of CVF1-4 at 25°C are less than 100 kPa, and are nearly zero at high temperatures, but do not lead to a viscoplastic state, indicating good pressure-sensitive properties.

DOI: 10.54254/2755-2721/91/20241091

Figure 5(c) shows that the loss factor-temperature curves also exhibit a similar trend: tan δ rises sharply at -30°C and -20°C, then gradually decreases. At 25°C, tan δ is much less than 1, and even when heated to 150°C, the proportion of viscosity does not increase, because hydrogen bonding between -COOH groups hinders chain mobility, contributing more to elasticity. Therefore, CVF primarily exhibits elasticity at room temperature, with overall stable viscoelasticity within the operating temperature range. The glass transition temperature (Tg) is the most crucial parameter for determining the minimum operating temperature of CVF; polymers exhibit viscous liquid state below Tg. The peak of the tan δ-T curve in Figure 5(c) corresponds to Tg, showing that all curves have only one glass transition temperature peak without significant peak width change, indicating the formation of a random copolymer among the reactants. The Tg values of CVF1-4 are all below -20°C, being -22.60°C, -24.94°C, -25.69°C, and -29.39°C respectively. Considering the Tg values of the copolymer monomers AA, 2-EHA, and n-BA are 106°C, -70°C, and -56°C respectively, according to the Fox equation (5-1):

$$\frac{1}{T_g} = \frac{W_1}{T_{g1}} + \frac{W_2}{T_{g2}} + \frac{W_3}{T_{g3}} + \cdots \ldots \tag{5-1}$$

The Tg of the copolymer can be predicted, decreasing with increasing graft content because the increase in graft structure dilutes the hydrogen bonds between AA components, enhancing molecular chain mobility. Additionally, pBA25 used for grafting is a soft segment with a low Tg, leading to a significant overall Tg reduction, making the product softer.

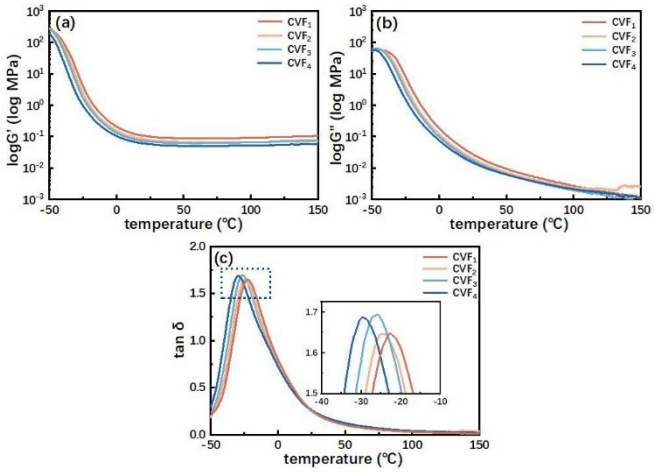


Figure 6. Temperature dependence of (a) storage modulus; (b) loss modulus; (c) loss factor of the grafted transparent viscoelastic body.

5.4. Continuous Tensile Cycles

Wearable electronics undergo significant bending and folding during use; therefore, their flexible substrates need to possess good reversible strain recovery to avoid buckling or wrinkling caused by residual strain. To characterize this performance of the acrylic CVF, this study examines the stress-strain relationship of the material during loading-unloading applications through continuous tensile cycles.

Overall, the room temperature cyclic process of four CVFs with different numbers of side chains exhibited reproducibility, as the curves obtained from ten consecutive experiments showed no significant differences. The maximum stress (stressmax) did not decrease at the maximum strain, and the stress at the initial strain point was almost at the original zero point, indicating that the prepared CVFs possess excellent recovery performance.

Figures 6(a), (b), and (c) respectively show the stress-strain curves of the four samples during the first, fifth, and tenth cycles. From these figures, it can be seen that with the increase in side chain content, the peak stress of the samples decreases sequentially. CVF1 has the highest peak stress, reaching 0.161 MPa, followed by CVF2 and CVF3 at 0.121 MPa and 0.0917 MPa, respectively. CVF4 has the lowest peak stress among the four, at 0.0675 MPa. As the number of short side chains composed of the soft

DOI: 10.54254/2755-2721/91/20241091

monomer BA increases, the Tg decreases, the material becomes softer, and its ability to resist stress diminishes.

In this study, the initial strain (Ɛn+1) at which stress begins to develop in the (n+1)th cycle is used as the residual strain for the nth cycle, according to equation (4-2).

$$R_n = \frac{300\% - \varepsilon^{n+1}}{300\%} \times 100\% \quad (5\text{-}2)$$

The strain recovery rates (R) for CVF1-4 during the first, fifth, and ninth cycles are shown in Table 3. CVF1-4 rarely exhibited yielding behavior, with strain recovery rates close to 100%. CVF1-4 nearly fully recovered in each loading-unloading cycle, with minimal irreversible breakage of weak bonds. Even after continuous cycling, the strain recovery rate only slightly decreased after nine cycles. For example, in CVF3, this value decreased from 95.46% to 95.05%, further demonstrating that CVF1-4 exhibit good elasticity within the testing range.

It was also found that CVFs, being viscoelastic materials, exhibit significant hysteresis behavior, with distinct differences between the loading and unloading stress-strain curves. This is due to energy dissipation caused by molecular chain friction during stretching, as well as hydrogen bonds and intermolecular forces within the CVFs. The hysteresis rate (H) reflects the sample's ability to recover elasticity and is related to the contribution of viscoelasticity: the greater the elasticity, the lower the hysteresis rate. CVF1 has the lowest hysteresis rate due to its fewer short side chains and higher density of intermolecular interactions and hydrogen bonds, demonstrating greater elasticity. The hysteresis rates of CVF2 and CVF3 are slightly higher and similar to each other, while CVF4 has the highest hysteresis rate, indicating a greater proportion of viscosity. As the number of cycles increases, the hysteresis rate of all samples decreases. Covalent bond breakage may occur in the initial cycle, but subsequent cycles mainly dissipate energy through hydrogen bonds or polar groups. This process is slow, and the properties tend to stabilize in subsequent cycles.

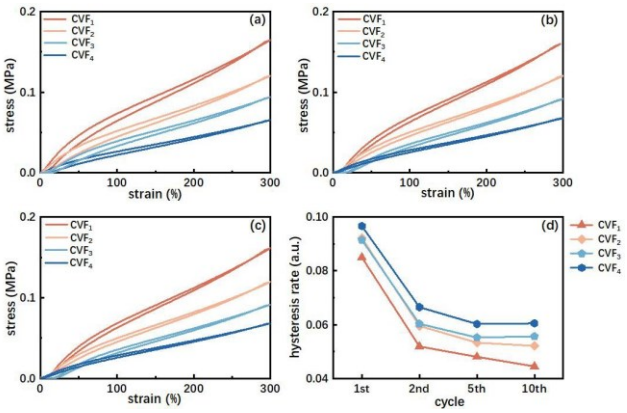


Figure 7. Grafted Transparent Viscoelastic Material (a) First Tensile Cycle (b) Fifth Tensile Cycle (c) Tenth Tensile Cycle (d) Hysteresis Rate

Table 3. Experimental Formulations

Sample	$stress_{max}$ (MPa)	R_1 (%)	R_5 (%)	R_9 (%)	H_1 (a.u.)	H_2 (a.u.)	H_5 (a.u.)	H_{10} (a.u.)
CVF_1	0.161	98.11	97.90	97.83	0.0850	0.0519	0.0480	0.0445
CVF_2	0.121	97.83	97.77	97.70	0.0922	0.0595	0.0533	0.0521
CVF_3	0.0917	95.46	95.39	95.05	0.0915	0.0603	0.0552	0.0556
CVF_4	0.0675	98.30	98.04	97.90	0.0966	0.0665	0.0602	0.0605

DOI: 10.54254/2755-2721/91/20241091

5.5. Research Summary

A series of grafted acrylic CVFs were successfully prepared through "grafting-through" and UV-initiated crosslinking. The products CVF1-4 consist of acrylic monomers, with short side chain structures imparting flexibility and viscosity. 2-EHA (Tg = -70 °C) and n-BA (Tg = -56 °C) provide primary flexibility, while the functional monomer AA (Tg = 106 °C) creates high-density hydrogen bonds. The addition of the crosslinking agent HDDA results in a gel rate of 93-97%, further enhancing the mechanical properties and stability of the samples.

To investigate the effect of grafting on viscoelastic properties, short side chains of pBA25 were introduced into the CVF backbone in mass fractions of 0%, 5%, 15%, and 30%, yielding four products: CVF1-4. At room temperature, their moduli are in the kPa range, with glass transition temperatures below -20 °C, and deformations reaching 500-600%, with nearly 100% reversible strain recovery. The products exhibit stable viscoelasticity within the -20~80 °C range, showing more elasticity at 25 °C. An increase in grafting quantity leads to softer CVFs, a decrease in glass transition temperature (from -22.60 °C to -29.39 °C), a maximum fracture strain of 572.442%, and a shear creep of 51.76%. However, the samples' Young's modulus, storage modulus, and loss modulus decrease at the same temperature, and the hysteresis rate of reversible tensile cycles increases, indicating that more side chains reduce elastic performance.

In summary, CVF3, with a moderate graft content, exhibits the best performance among the four samples, showing good and balanced viscoelasticity within the operational temperature range.

6. Conclusion

High-transparency acrylic CVFs with clear short grafted structures and high-density crosslinked networks were successfully synthesized based on the free radical reaction mechanism. The random copolymer products CVF1-4 exhibit a single, low glass transition temperature, making them suitable for a wide temperature range. As viscoelastic materials, they possess good and enduring stable mechanical properties, capable of multiple deformation and recovery cycles, and rarely exhibit irreversible damage under large deformations.

Due to different graft content ratios and similar gel rates, CVF1-4 display unique mechanical properties. Overall, as the short side chains of pBA25 increase, the density of functional carboxyl groups capable of forming hydrogen bonds decreases, making the acrylic CVFs softer. Although they can undergo greater deformations, their ability to recover reversible strain declines, and they struggle to resist significant stress.

In this study, the modulus decrease caused by increased side chains seems highly influenced by the copolymer composition, varying with different circumstances, and requires further research. For example, although pBA25 as a side chain structure expands the molecular chain volume and can dilute intermolecular hydrogen bonds and other forces, reducing cohesive forces by lowering crosslinking density, CVF3, containing 15% side chains, shows the highest fracture stress and initial adhesion, possibly due to its slightly higher gel rate compared to the other three, having a more significant effect in these aspects than crosslinking density. Additionally, the overall performance difference between CVF1 without grafting and CVF2 with 5% side chains is minimal, possibly because the BA monomer itself has shorter and smaller side chain groups, which are less effective in reducing chain entanglement and friction caused by hydrogen bonds compared to copolymer monomers like 2-EHA with larger side groups [13]. BA homopolymers are more prone to entanglement than BA-EHA copolymers of the same molecular weight. Adding more BA monomers might reduce the mobility of polymer chains, but no definitive studies have yet clarified the relationship between the two.

An ideal CVF should have high fracture strength to avoid potential damage in practical applications and a large fracture elongation to accommodate significant deformations during folding. Therefore, to achieve CVFs with excellent comprehensive performance, an appropriate molecular composition and structure should be selected to balance its mechanical properties, such as viscosity and elasticity, ultimately enabling applications in flexible sensors.

DOI: 10.54254/2755-2721/91/20241091

References

[1] Qian Xin, Su Meng, Li Fengyu, et al. Advances in Research on Flexible Wearable Electronic Sensors [J]. Journal of the Chinese Chemical Society, 2016, 74(07): 565-575.

[2] Jin Bin, Zhang Peng, Xia Jianhui. Study of Acrylic Optical Transparent Viscoelastic Materials for Flexible Displays [J]. Adhesion, 2022, 49(06): 1-6.

[3] Czech Z. Synthesis and cross-linking of acrylic PSA systems[J]. Journal of Adhesion Science and Technology: The International Journal of Theoredtical and Basic Aspects of Adhesion Science and Its Applications in All Areas of Technology,2007,21(7):625-635.

[4] Mitchell M, Erfan D, N A K, et al. Sticky Architecture: Encoding Pressure Sensitive Adhesion in Polymer Networks. [J]. ACS central science,2023,9(2):197-205.

[5] Sylwia D ,Francesca L ,Adam G , et al. Toward Green Atom Transfer Radical Polymerization: Current Status and Future Challenges. [J]. Advanced science (Weinheim, Baden-Wurttemberg, Germany),2022,9(19):e2106076-e2106076.

[6] Baek S, Jang S, Hwang S. Preparation and adhesion performance of clear acrylic pressure sensitive adhesives: effects of substituent structure of acrylate monomer[J]. International Journal of Adhesion and Adhesives,2016,64:72-77.

[7] Lee H J, Myung H M, Baek J M, et al. Effects of monomer functionality on physical properties of 2-ethylhexyl acrylate based stretchable pressure sensitive adhesives[J]. Polymer Testing,2019,76:305-311.

[8] Lee J, Lee T, Shim K, et al. Molecular weight and crosslinking on the adhesion performance and flexibility of acrylic PSAs[J]. Journal of Adhesion Science and Technology, 2016, 30(21): 2316-2328.

[9] Ingale P R, Jagtap N R, Adivarekar V R. Effect of Molecular Weight on Performance Properties of Pressure-Sensitive Adhesive of Poly (2-ethylhexyl acrylate) Synthesized by RAFT-Mediated Miniemulsion Polymerization[J]. The Journal of Adhesion,2016,92(3):236-256.

[10] Wang W, Lu W, Goodwin A, et al.Recent advances in thermoplastic elastomers from living polymerizations: Macromolecular architectures and supramolecular chemistry[J].Progress in Polymer Science,2019,95:1-31.

[11] Anastasia M, Wenwen L, Robert G, et al. Soft Elastomers via Introduction of Poly (butyl acrylate) “Diluent” to Poly (hydroxyethyl acrylate)-Based Gel Networks[J]. ACS Macro Letters, 2013, 2(1):23-26.

[12] Heeyeon M, Kihoon J, Jin M K, et al.Solvent-free Deposition of Ultrathin Copolymer Films with Tunable Viscoelasticity for the Application to Pressure Sensitive Adhesives.[J].ACS applied materials interfaces,2018,10(38):32668-32677.

[13] Dongdong W, Jiapeng L, Fushi L, et al. Solvent-free and biodegradable pressure-sensitive adhesive based on chlorosulfonated Poly (propylene carbonate) [J]. Polymer, 2023, 286: 126418-126418.