



Layer-by-layer assembled transparent polymeric adhesive films with adjustable refractive indices

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ABSTRACT

Materials with high adhesive strength that can be fabricated by environmentally friendly methods are crucial for various bonding applications. In this study, highly adhesive films with an adjustable refractive index (RI) were prepared by the layer-by-layer (LbL) assembly of cationic poly(diallyldimethylammonium chloride) (PDDA) and an anionic blend of poly(acrylic acid) (PAA) and poly(4-styrenesulfonic acid) (PSS). The RI of the adhesive films can be precisely controlled between 1.5181 and 1.5562 by simply changing the PSS content. We obtained high adhesive strengths in the range 3.99 ± 1.04 MPa to 6.88 ± 0.95 MPa by gently pressing two glass substrates together with transparent (PDPA/PAA-PSS)*n LbL assembled films with different NaCl contents in the presence of a thin layer of deionized water (n refers to the number of film deposition cycles). Importantly, the water is an environmentally friendly solvent in the bonding and debonding processes. Finally, these adhesive films exhibit high optical transparency in the visible region and exhibit excellent thermal stability. The outstanding features of these films ensure their use in practical optical bonding applications.

1. Introduction

The demand for optically clear adhesives for optical bonding applications is increasing as the optics industry matures. Robust optical bonding with high transparency is essential for advanced optical systems such as touch panels and display devices [1–3]. The bonding of glass components improves the mechanical durability of optical systems. Moreover, optical bonding can effectively improve the image quality of optical displays because it reduces the amount of reflected light by eliminating the air gap between bonded glass components, this increases the contrast ratio. However, matching the RI of the adhesives with the glass components is required in such applications because an overly large difference in the RI values will negatively affect the image quality due to light scattering at the interface. Therefore, the RI of an optical adhesive must be adjusted to match the RI of the optical components at hand [4,5]. Moreover, adhesive films, compared with paste, with homogeneous surfaces capable of eliminating the bubbles can be directly applied to optical bonding applications [3,6]. Traditional adhesive films, some involving frequently heating or pressing processes

limit the application in assembly of precise optical components [3]. Therefore, adhesive films with adjustable RI which can fit a non-planar substrate, direct surface adsorption on a substrate, and possess controlled thickness are highly desirable.

Numerous methods have been developed to tailor the RI of polymer matrices such as the hybrid methods and the introduction of high-RI moieties [7]. Polymer-inorganic hybrid composite materials have attracted great interest in recent years due to their outstanding optical properties. The use of high-RI inorganic nanoparticles in polymer systems results in high-RI polymer-inorganic composite materials. However, hybrid polymer-inorganic composites lack mechanical flexibility, which is an essential characteristic for optical adhesives. In contrast, increasing the RI of polymeric matrices is a promising approach for designing optical adhesives. Introducing high RI moieties such as aromatic rings, sulfur, and heavy halogens can increase the RI of a polymer system.

Previous work has yielded interesting results. For example, Ueda and co-workers [7–9] reported a series of polyimide compounds with sulfur-containing groups having RI values greater than 1.7. Matsumoto

Abbreviations: RI, refractive index; LbL, layer-by-layer; PDPA, poly(diallyldimethylammonium chloride); PAA, poly(acrylic acid); PSS, poly(4-styrenesulfonic acid); n, the number of film deposition cycles; PVS, poly(vinyl sulfate) potassium salt; UV, Ultraviolet; UV-vis, Ultraviolet-visible; SEM, Scanning electron microscopy

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et al. [10] employed poly ring compounds to synthesize alicyclic polyimide through a two-step synthesis process and then prepared polyimide films with different contents of aromatic compounds with RI values of 1.522–1.617. However, high RI polymeric materials with adhesive functions are rare. To obtain optical adhesives with the desired RI properties, an easy processing method involving a high-RI polymer is important. This suggests the use of a commercially available water-soluble polymer containing benzene and sulfur. These conditions can be achieved with PSS.

LbL assembly is an important technique for preparing multi-functional and multilayer films via the alternate deposition of film layers on solid substrates. The substrate is alternately immersed in a different polyelectrolyte solution for an appropriate time then washed to remove the physically adsorbed polyelectrolytes [11–15]. The thickness, composition, and structure of LbL assembled polyelectrolyte films can be controlled by adjusting the parameters of the polyelectrolyte solution such as concentration, pH, and so on [11,13,14]. Multilayered LbL assembled films with desired functionalities have been successfully prepared via numerous driving forces [16,17]. Multilayer LbL assembled films include numerous building blocks such as synthetic macromolecules, particles, small molecules, and biomacromolecules [11–13,18]. The LbL technique has numerous advantages in the preparation of functional films. First, it offers uniform deposition of films on the substrates with non-flat surfaces and arbitrary shapes and sizes. Second, the film preparation process is simple and requires no complicated equipment. Third, it is a facile technique to build multi-component films requiring accurate control of film composition and thickness. Therefore, the LbL assembly technique is a promising method for the preparation of polymer films with both optical and adhesive functionalities. However, the rapid preparation of LbL assembled adhesive films using PDDA and PSS films with larger thicknesses remains challenging because of the repulsive interactions between the strongly charged PDDA and PSS [19]. Thus, many assembly processes are necessary to prepare thicker films. Nevertheless, the deposition efficiency of such films can be increased by introducing NaCl into the polymer system [19–21]. This is because the molecular chains of polymers become more coiled with increasing NaCl concentration resulting in greater film thickness. This was demonstrated by Kim and co-workers [20] in 1999 when NaCl was introduced into a poly(vinyl sulfate) potassium salt (PVS) and PAA system. They then reported the thicknesses and surface morphologies of the films. The results showed that the film thicknesses were greater than a corresponding system without NaCl under equivalent conditions. The effect of NaCl addition on the infrared absorption of the films was negligible. Goh and co-workers [21] studied the effect of NaCl concentration on the thickness and other characteristics of (PDDA/PSS)**n* LbL assembled adhesive films comprising *n* PDDA/PSS layers. The thickness of the adhesive films showed an approximately linear increase with increasing NaCl concentrations up to 1 M for a given value of *n*. In 2009, Serizawa et al. [22] fabricated LbL assembled adhesive films using PDDA and PSS solutions with a 1 M NaCl concentration on silicon wafers and polyimide substrates. The results demonstrated rapid film growth due to the addition of NaCl. Moreover, the films could be bonded by adding water to the contact surface.

Ultraviolet (UV)-curable optical adhesives, cyanoacrylate, and epoxies are the most widely used adhesives in optical-bonding processes [1,2,5,23,24]. The primary advantage of UV-curable adhesives is that the bonding process can be completed within a few seconds under UV light. However, the debonding process of these adhesives generally requires the use of heat treatment or immersion in organic solvents such as acetone and dichloromethane [5,25,26]. The heating process can deform the optical components, and the use of organic solvents can be detrimental to the optical components and hazardous to personnel. In addition, the curing process of epoxies can be sensitive to oxygen and water in the air, and thus the components must be bonded very quickly after the application of the adhesive [3]. Therefore, it is critical to

develop adhesive films with a continuously adjustable RI that are insensitive to atmospheric conditions and are benign to people, the environment, and other optical components.

To address these vital concerns, this report presents the preparation of transparent and highly adhesive films via LbL assembly using aqueous solutions of PDDA and a blend of PAA-PSS with different NaCl contents with adjustable RI, as well as acceptable bonding strengths. LbL is critical in this work because the adsorption on a rough substrate led to a smooth surface, which avoided the formation of bubbles. Moreover, adhesive films with controlled thickness and composition were homogeneously deposited. The adhesive films are highly transparent in the visible region with an adjustable RI. Here, NaCl was introduced to the system as a typical inorganic salt to change the ionic strength and both enhance the film thickness and, more importantly, produce a transparent film that benefits optical applications. The RI of the polymer system can be adjusted by simply changing the PSS content in the PAA-PSS aqueous solution. The resulting (PDDA/PAA-PSS)**n* adhesive films deposited on substrates can be strongly bonded by gentle pressure in the presence of a thin water layer (*n* refers to the number of film deposition cycles). They exhibit sufficient thermal stability for practical applications. Furthermore, debonding can be easily accomplished by quickly immersing bonded components in HCl. The resulting homogenous and smooth (PDDA/PAA-PSS)**n* adhesive films eliminate the bubbles and can be directly applied to optical bonding applications. The combined benefits of the proposed adhesive films offer significant potential for numerous optical applications.

2. Materials and methods

2.1. Materials

PDDA (Mw ca. 100,000–200,000), PAA (Mw ca.1800) and PSS (Mw ca.75000) were purchased from Sigma-Aldrich. No.7148 AB Epoxy adhesive was purchased from Deli group. Lantian 9301 UV curing adhesive was purchased from Shenzhen Special New Material Co., Ltd. All chemicals were used without further purification. Deionized water was used for all the experiments.

2.2. Methods

2.2.1. Preparation of (PDDA/PAA-PSS)**n* adhesive films

PDDA solution with different NaCl content was prepared as follows. PDDA (0.1 g) was dissolved into deionized water (100 mL). Then different contents of NaCl were added into the PDDA solution, as shown in Table 1. The PAA-PSS aqueous solution was obtained by the following procedure: PAA and PSS (a total of 0.1 g) were added to deionized water (100 mL). Then different contents of NaCl (0 mol , $4.60 \times 10^{-2} \text{ mol}$, $7.16 \times 10^{-2} \text{ mol}$, $9.22 \times 10^{-2} \text{ mol}$ and $1.00 \times 10^{-1} \text{ mol}$, respectively) were added into different molar ratios of PAA-PSS (1:0, 3:1, 1:1, 1:3 and 0:1, respectively) aqueous solution correspondingly. The pH values of PDDA and PAA-PSS solution were adjusted to 8 and 3.5 using 1 M NaOH and 1 M HCl, respectively. Solution compositions of combinations of PDDA and PAA-PSS are also listed in Table 1. Glass substrates were sonicated in anhydrous ethanol for 30 min to remove

Table 1
Solution composition in the combination of PDDA and PAA-PSS.

PDDA [1 mg mL^{-1}]		PAA-PSS [1 mg mL^{-1}]	
PDDA [g]	NaCl [g]	PAA-PSS [molar ratio]	NaCl [mol]
0.1	0	1:0	0
0.1	5.85	3:1	4.60×10^{-2}
0.1	5.85	1:1	7.16×10^{-2}
0.1	5.85	1:3	9.22×10^{-2}
0.1	5.85	0:1	1.00×10^{-1}

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