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Strong visible-light emission in annealed poly(acrylic acid)

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Abstract: An intense and broadband photoluminescence emission is demonstrated in a bio-compatible film produced by thermal annealing of poly(acrylic acid) (PAA). The annealing process enhances the emission intensity from weakly emitting sub-luminophores of C–O and C=O functional groups by a factor of more than two orders of magnitude. The emission property can be sensitively tuned by the annealing temperature, duration, the PAA solution preparation conditions such as solvent type and solvent:PAA ratio. The strong luminescence is attributed to the formation of rigid molecular structure due to aggregation and crosslinking. FTIR measurements show that the aggregation and crosslinking processes subdue nonradiative recombination pathways.

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1. Introduction

Luminescent materials in different forms continue to garner research attention due to their wide range of applications. Organic emitters are attractive for biotechnology and development of sustainable rare-earth-free light sources. Conventional luminescent organic molecules contain aromatic functional groups with extended \( \pi \)-conjugation. This class of emitters are often toxic and suffer from aggregation-induced quenching [1]. Recently, non-conventional, aggregation-induced emission (AIE) in non-conjugate molecules containing weakly emitting functional groups, known as sub-luminophores, such as C=O, C=N, N=O, and C–O, emerge as a new class of luminescent materials [2–5]. For example, visible light emission based on the C=O functional group has been observed in poly[(maleic anhydride)-alt-(vinyl acetate)] (PMV) solution [6]. Aggregation and cross-linking processes promote radiative recombination by suppressing nonradiative recombination pathways, leading to high quantum yield [7]. The new AIE concept provides a new pool of materials for the development of heavy-metal-free, non-toxic and biocompatible light sources for in vivo biomedical applications.

A comprehensive review by Mei et al. showed extensive molecular engineering of a variety of advanced organic molecular structures in great depth for emission control and a wealth of potential applications [8]. To harness their potential, full understanding of the complex mechanisms of aggregation, crosslinking, radiative and non-radiative transitions is relevant. In this regard, less-complicated organic molecules are useful. Poly(acrylic acid) (PAA) can be a suitable candidate. Acrylic acid is a simple carboxylic acid molecule that contains two weakly emitting sub-fluorophores: C=O and C–O.

Poly(acrylic acid) is a benign organic molecule, which is used widely for industrial applications such as disposable diapers, detergents and adhesives. Furthermore, it attracted limited optoelectronic applications such as for the formation of fluorescent and photoresponsive supramolecular polymer gel [9], for the passivation of the fluorescent-porous silicon [10], as cross-linking agent in fluorescent label preparation in cancer cell imaging [11], and for surface functionalization by grafting for preparation of fluorescent-based bioassays [12]. Recently, clustered-triggered emission in amorphous PAA is observed where strong NUV phosphorescence in solution and solid-state forms [13]. Owing to its content of the weakly emitting sub-luminophores, PAA has a potential to be an efficient luminescent organic polymer.
In this article, we report a broadband visible photoluminescence from a film produced by annealing of PAA without involving other organic emitters. We show that PAA can be processed to be an efficient luminescent material without advanced chemical processing. Characteristics of the PL emission property achieved by different annealing conditions is discussed. FTIR measurements reveal that strong emission is induced by aggregation and crosslinking processes. AIE investigations are often performed in solutions, where the aggregation process is achieved by reducing solvent concentration or in powder forms. Most industrial applications, such as displays and LED’s, require film-based production of the light source. Therefore, it is essential that the AIE emitters are prepared in a form of solid films, which is achieved in this work.

2. Experiment

White PAA powder with a molecular weight of 1800 g/mol is bought from Sigma-Aldrich. The powder is dissolved in either deionized water or ethanol. The ethanol is of high purity (99.8%), and it is also bought from Sigma-Aldrich. Ratios between the solvents (water, ethanol) and PAA was maintained at either 3:1 or 4:1 ratio by weight. The ratios mentioned hereafter are by weight. The mixture of the solvent and the PAA is stirred with a magnet stirrer until the PAA dissolves completely, which takes approximately 30 minutes. Then, small pieces of wafers [silicon, silicon carbide and microscope slide (borosilicate glass)] are placed inside the solution for 20 minutes. Afterwards, the sample is placed in a fume hood and left to dry in ambient atmosphere overnight. Dry and transparent films are formed on the substrates.

To achieve an intense visible photoluminescence, annealing is conducted by placing the samples on a hot plate at different temperatures between 200–400 °C and durations. The annealing process is conducted in ambient environment.

Micro-PL setup in a front illumination/light collection setting, using a single microscope objective, is used to characterize the emission property of the annealed PAA films. The schematic of the measurement setup is shown in the inset of Fig. 1. Continuous wave lasers (center wavelength at 375 nm or 405 nm) are focused to a spot diameter of approximately 110 µm for the excitation of the sample. The incident power is set at 150 µW unless specified. The PL signal is collected by the microscope objective and is fiber-coupled to an optical spectrometer (SR-303I-A spectrograph with DU-420-0E CCD). The PL signal is transmitted through a long-pass filter with a cut-off wavelength of 405 nm. The CCD camera is cooled to a temperature of −70 °C to reduce background noise.

Time-resolved PL (TRPL) measurements are conducted with a time correlated single photon counting (TCSPC) system from PicoQuant GmbH. The micro-PL setup is also used for the TRPL measurements. A picosecond laser (375 nm) with a pulse width of approximately 44 ps is used for the excitation of the sample in the TRPL measurements. The PL signal is fiber coupled into a single photon detector (PMA Hybrid from PicoQuant GmbH).

Finally the Fourier transform infrared spectroscopy (FTIR) measurements are conducted by a FTIR spectrometer (VERTEX 70, Bruker) in a transmission mode with a resolution set at 2 cm⁻¹. For the FTIR measurements presented in the main article, the PAA is deposited on Si substrates. The Si substrates have low absorption in the measurement spectral range (500–4000 cm⁻¹) and the PAA’s FTIR fingerprints are not obscured by the substrate absorption in the transmission mode measurements.
3. Results

During the annealing process, volatile gas byproducts are observed by naked eyes. The annealing temperatures are above the boiling point of water and water is expected to be a gaseous byproduct. It has previously been shown that thermal annealing of PAA at 170–440 °C leads to volatile byproducts and new chemical structures such as anhydrides and cold ring fractions can be produced [14]. When PAA is heated above 200 °C, it crosslinks in the dehydration process to form a biodegradable poly(acrylic anhydride) polymer [15]. Note that poly(anhydride) polymer is a useful biomaterial for drug delivery to various organs [16]. The partial conversion of the PAA to poly(anhydride) in the annealed samples is confirmed by FTIR measurements and is discussed in more detail later. The annealing process affects the color of the PAA films. For example, for 5 minutes annealing at temperatures of 200 and 250 °C, the PAA films are transparent. The film turns brown at 400 °C-annealing. For the annealing temperature of 400 °C, significant chain breakdown can be expected [15]. The annealing duration also affects the color of the film. The PAA film stays transparent for 1-minute annealing at 350 °C annealing. A 2-minutes annealing at 350 °C results in a yellowish color. The photograph images of PAA films with different

![Fig. 1. PL emission from annealed PAA on a SiC substrate: (a) emission spectra with 375 nm-laser excitation, (b) transient of PL decay and the instrument response function (IRF) with 375 nm-pulsed-laser excitation. The fitting parameters of the PL decay with a three-phase exponential decay is included as an inset. The PAA film is annealed at 350 °C for a period of 5 minutes. The inset in Fig. 1(a) shows schematic of a micro-PL setup used for the characterization of PL emission. DM stands for dichroic mirror.](image-url)
annealing conditions are shown in the Supplement 1 [see Fig. S1(a)] The yellowish color can be a qualitative indicator for the efficient luminescent layers. The PAA films have relatively smooth surfaces as shown by SEM images [see Fig. S1(b)] in the Supplement 1. The films show cracking signs due to bad mechanical adhesion with the substrate which can be improved by making the film on porous SiC substrates.

Figure 1(a) shows the PL emission spectra of annealed PAA film on a silicon carbide (SiC) substrate. The PAA is annealed for a period of 5 minutes at a temperature of 350 °C. SiC is an optical material with exceptional material properties, suitable for operation under high thermal load [17,18] and it is used industrially as a substrate for light emission. The emission from the SiC substrate, without the luminescent PAA, is included in Fig. 1(a) for reference. The emission from not-annealed PAA on the SiC substrate is similar to that of the SiC substrate emission. This clearly shows that the luminescence efficiency is significantly improved by annealing and without advanced chemical processing steps.

To characterize the emission process further, TRPL measurements of the PAA film on SiC substrate are conducted, and PL decay curves are shown in Fig. 1(b). Additional TRPL measurements of PAA films on glass substrates are shown in the Supplement 1 (see Fig. S2). The transient PL decay is fitted with a three-phase exponential function, \( f(t) = A + B_1 \exp(-t/\tau_1) + B_2 \exp(-t/\tau_2) + B_3 \exp(-t/\tau_3) \) where \( A, B_1, B_2 \) and \( B_3 \) are constants, and \( \tau_1, \tau_2 \) and \( \tau_3 \) are the lifetimes for different recombination pathways. The average carrier lifetime, calculated from \( \tau_{av} = \sum_i B_i \tau_i^2 / (\sum_i B_i \tau_i) \), where \( i = 1–3 \), is 3.34 ns and it is a small value, which is typical of efficient light converters [19]. The three exponential decay functions imply more than one radiative recombination pathways. The presence of two sub-luminophores leads to two different recombination pathways. The third phase in the exponential fit could be related to the non-radiative recombination pathway.

To investigate the emission mechanism and tunability in more detail, the annealing temperature and duration for PAA film on high resistivity Si (HRSi) and glass substrates are varied. The emission spectra for PAA film on glass substrate are shown in Fig. 2 and those of PAA on HRSi are shown in the Supplement 1 (See Figs. S3 and S4). Strong PL emission, where the emission intensity is enhanced by more than two orders of magnitude by annealing, is observed from both sets of samples. Similar emission property from both sets of samples, irrespective of the substrate, shows that the strong emission is from the PAA film and not from PAA-substrate interface complex.

Figure 2(a) shows that emission intensity and spectra can be tuned by varying the annealing temperature and thereby the film chemical structure. An annealing duration of 5 minutes is used for samples characterized by the measurement results in Fig. 2(a). As we increase the annealing temperature from 200 to 300 °C, the PL intensity increases dramatically. The integrated PL intensity over a wavelength range of 400–700 nm increases by factors of 17 and 13 for annealing temperatures of 300 and 350 °C relative to the annealing temperature of 200 °C. Annealing temperature of 400 °C results in a drastic decrease in the emission intensity, where the integrated PL intensity decreases by a factor of 4 relative to that of 200 °C-annealing temperature. This is due to significant chain breakdown and material degradation [15]. Furthermore, as we increase the annealing temperature, the PL spectra shift towards longer wavelengths as depicted in the normalized emission spectra of inset of Fig. 2(a). As annealing temperature changes from 200 °C to 400 °C, the emission peak shifts from 439 nm to 562 nm. This can be attributed to the changes in the size of the aggregation cluster as well as changes in the contributions of different luminescent species. A stronger aggregation results in stronger intra- and inter-chain interactions, which in turn leads to red-shift of the emission spectrum [2]. The presence of two sub-luminophores in C=O and C–O indicates that there could be at least two radiative emission mechanisms based on each sub-luminophore where the relative contributions to the radiative recombination can depend on the annealing temperature.
Fig. 2. Impact of annealing temperature and duration on the emission property of annealed PAA on borosilicate glass. (a) PL emission spectra at several annealing temperatures for a constant annealing duration of 5 minutes. The PAA film in (a) is made from a solution of ethanol-PAA mixture in 4:1 ratio. The inset in (a) is plot of the normalized PL spectra. (b) PL emission spectra at different annealing conditions. The PAA film in (b) is made from a solution of water-PAA mixture in 3:1 ratio. The inset in (b) shows emission spectra at different excitation exposure durations. The other PL measurements are done immediately after excitation exposure (short exposure time).
The emission properties can also be tuned by annealing duration. Measurements of the PL emission from annealed-PAA film on glass substrate are shown in Fig. 2(b). The PAA solution is made by mixing PAA powder and water in 1:3 weight ratio. We consistently note that increasing the annealing duration at a given temperature shifts the emission spectra to longer wavelength. Increasing the annealing duration from one to 2 minutes increases the emission intensity significantly. Further increase of annealing duration to 5 minutes, results in weaker but broader emission spectra, centered at approximately 600 nm. The integrated PL intensity over the wavelength range of 400–700 nm, increases by 34% for annealing duration of 2 minutes and decreases by a factor of 159 for annealing durations of 5 minutes relative to that of the 1 minute-annealing duration. The weak emission for the 5 minutes-annealing duration is attributed to chain breakdown and material degradation. Annealing for 5 minutes results in degradation of the luminescent complex. We, interestingly, observe that this sample shows photoresponsivity, where the emission property can be modified by the laser excitation. The emission intensity tends to increase with the excitation-laser exposure time as shown in the inset of Fig. 2(b). With increase in the exposure time by up to 12 minutes, the PL intensity increases and the emission spectrum blue-shifts. This shows that laser exposure can be used in the optimization of the emission property. The observed photoresponsivity can also have many potential uses in biomedical applications such as biosensing [9,20–23].

The measurements in Fig. 2 clearly demonstrate a significant enhancement in the emission intensity by more than two orders of magnitude in the annealing processes. The quantum yield of the PAA film annealed at 350 °C for 2 minutes is estimated to be 8.9%. The PL spectrum of this sample is the water blue curve in Fig. 2(b). The carrier lifetime of the PL decay is 3.3 ns. The details of the quantum yield evaluation are included in the Supplement 1 (Fig. S5).

The compositions of the PAA solutions used in the measurements of Figs. 2(a) and 2(b) have different weight ratios of PAA and water (1:4 and 1:4 respectively). This is intentionally made to see the impact on the PL emission property. The annealing and PL measurement conditions of the pink curve in Fig. 2(a) and the red curve in Fig. 2(b) are the same. However, both the emission intensities and spectra are significantly different where the later results in a weaker intensity by more than two orders of magnitude. The PL peak of the pink curve in Fig. 2(a) is 504 nm and it red-shifts to 580 nm for the red curve in Fig. 2(b). This shows that several parameters of the film preparation and annealing conditions affect the emission property sensitively.

We note that the annealed PAA does not have strong adhesion with flat Si, SiC and glass substrates and it peels off easily. Deposition of PAA on porous SiC surface improves the mechanical adhesion. The porous layer is fabricated by an anodic oxidation process [24,25]. The PL emission from the PAA on a porous SiC is shown in Fig. 3. To clarify the possibility of exciting the emission with lasers at longer wavelengths, PL spectra are also measured using a 405 nm-laser excitation. The 405 nm-laser excitation results in PL emission which red-shifts in comparison to the 375 nm excitation. This shows that different radiative recombination pathways are preferentially invoked by the different excitation wavelengths. Relative comparison between the conversion efficiencies of the two spectra, by integrating the PL spectra, shows that 375 nm excitation is approximately 38% more efficient than 405 nm excitation. The excitation-wavelength dependence can be used for wavelength tuning and the possibility to use the longer-wavelength excitations is suitable for practical applications as the longer-wavelength blue lasers are industrially more accessible.

In order to clarify the physical mechanism of the emission enhancement, FTIR measurements are performed. Figure 4 shows the FTIR transmission spectra of annealed PAA deposited on HRSi substrates with different annealing conditions. The PL emission properties of these samples are shown in the Supplement 1 (Figs. S3 and S4). The PAA solution for the measurement results of Fig. 4(a) contains ethanol and PAA powder in 4:1 weight ratio. The PAA layers of the samples used for the measurement results in Fig. 4(b) are made from PAA-water solution in 1:4 ratio.
Fig. 3. PL emission spectra of annealed PAA on porous SiC with 375 and 405 nm excitation lasers. The annealing temperature is 350 °C and the duration is 5 minutes. The PL signal is recorded immediately after excitation exposure (short exposure duration).

Fig. 4. FTIR transmission spectrum of PAA deposited on HRSi substrates at different annealing conditions. The PAA layer is made of (a) ethanol solution, and (b) water solution. The solvent to PAA ratio is 4:1 and the annealing duration is 5 minutes. The green curve in Fig. 1(b) is rescaled for visual clarity. The plot section enclosed by the dashed rectangle is magnified in the inset of Fig. 1(a) for clarity. The curves in this inset are normalized by their values at 2000 cm$^{-1}$. The insets in Fig. 1(b) shows a ball-and-stick model of acrylic acid (right) and acrylic anhydride (left). The absorption bands of relevant functional groups in the discussion are shown by arrows.
The C=O, C–O, and O–H bonds in PAA give important fingerprints in the infrared spectral region. We observe a strong absorption band centered around 3300 cm\(^{-1}\) and extending approximately from 2400 cm\(^{-1}\) to 3600 cm\(^{-1}\) in both annealed and not-annealed samples. The O–H stretch is the source of this absorption feature and the broadening is due to strong intermolecular interaction, mainly from the hydrogen bonding. The large absorption bandwidth is a common feature of other molecules containing O–H bonds such as water and carboxylic acids [26]. The other prominent FTIR features from the C=O stretch and C–O stretch are expected at approximately 1700–1730 and 1210–1320 cm\(^{-1}\), respectively [27]. Experimentally, we observed strong absorption bands around 1610–1760 cm\(^{-1}\) and 1130–1310 cm\(^{-1}\) as shown in Fig. 4. Several other weaker transmission dips are also observed on the lower wavenumber side of the measurement range. Detail assignments of the peaks have been reported previously [28,29].

Different FTIR spectral features appear after annealing of the PAA layer. Here, we discuss the important features that are relevant for the explanation of PL observations. Firstly, the strength of FTIR signatures due to the functional groups is reduced. For example, the O–H absorption band gets narrower and weaker. At high temperature annealing, PAA changes into poly(acrylic anhydride) with the release of water molecules. As can be seen in the inset of Fig. 4(b), acrylic anhydride does not contain the O–H functional group. However, the persistence of the strong O–H band after annealing indicates the poly(acrylic acid) is not completely transformed into poly(acrylic anhydride). Therefore, the measured spectra of the annealed PAA contain contribution from both PAA and poly(acrylic anhydride). Secondly, a new spectral dip appears near 2941 cm\(^{-1}\). This is due to C–H stretching vibrations [28]. These absorption signals are less obscured by the O–H absorption band after annealing.

Thirdly, a new feature centered around 1804 cm\(^{-1}\) appears. This feature does not appear in the not-annealed PAA FTIR transmission spectra. This is related to C=O stretch and presence of poly(acrylic anhydride) [30]. In acid anhydrides, two C=O bonds are connected by an oxygen bridge as shown in the inset of Fig. 4(b). Coupling between the two C=O bonds introduces two nearby IR fingerprints due to in-phase and out-of-phase stretching motions. The peak at 1804 cm\(^{-1}\) can thus be attributed to symmetric C=O stretch which is expected to appear at higher energy than the asymmetric C=O stretch. The asymmetric stretching mode is expected approximately at 1750 cm\(^{-1}\). The single C=O stretch in PAA is expected at smaller energy (approximately 1700–1730 cm\(^{-1}\)) than the two coupled modes. As can be seen in the inset of Fig. 4, the C=O band near 1700 cm\(^{-1}\) narrows, shifts to higher energy and weakens as the annealing temperature increases, which is consistent with the partial conversion of the PAA into the poly(acrylic anhydride). Similarly, a strong absorption band is observed near 1250 cm\(^{-1}\) due to the C–O stretching motion. In the annealed sample, the absorption strength decreases and the band appears as absorption dips near 1175 and 1275 cm\(^{-1}\).

The absence of the absorption peaks near 1804 and 2941 cm\(^{-1}\) in the not-annealed PAA are difficult to see in Fig. 4 due to the strong absorptions bands by C=O and O–H units where the transmission is significantly suppressed. By measuring a thin layer of not-annealed PAA on an n-doped Si wafer, which allows non-zero transmission near these bands, we show that 1804 and 2941 cm\(^{-1}\) are not observable in the not-annealed PAA (see Fig. S6 in the Supplement 1).

We note that films made from PAA solutions of both water and ethanol lead to strong photoluminescence emission by annealing (see Figs. S3 and S4 in the Supplement 1). Comparing the transmission spectra of PAA from ethanol and water solutions, it can be observed that prominent features of the three functional groups and temperature-dependent trends are similar. More features are apparent in the ethanol solution at low frequencies from the ethanol traces such as C–C–O symmetric and unsymmetrical stretching motions, and O–H out-of-plane and in-plane bending motion [31].
4. Discussion

It is clear from the experimental observations that annealing of the PAA leads to weakening of infrared absorption by the C=O, C–O, and O–H bonds, which is correlated with improvement in the PL emission intensity. The reduced absorption means that the molecular vibrations and rotations are reduced. The aggregation process induces rigid molecular structure through the formation of new conformations, new covalent bonds, stronger intra/inter-molecular long-range bonds, molecular entanglement, and strong electrostatic interactions among the organic molecular units, which inhibit the non-radiative decays. As a result, quantum yield of the light emission improves. The FTIR measurement allows direct observation of suppression of the molecular vibrations in the aggregation-induced process. The aggregation process, leads to overlap of intra- and intermolecular electron clouds. This forms extended conjugation of n-π* and π-π* interactions which improves the luminescent efficiency [32]. Aggregation also influences the emission spectra. It reduces the distance between functional groups, which, in turn, leads to wave functions of the electron clouds to overlap and couple. This results in the reduction of the gaps between energy levels and creation of new energy levels [3]. This affects the absorption property of the non-conjugate polymers. We observed that the strong emission of the thermally annealed PAA is characterized by strong absorption (13% transmission) of the excitation light. Before annealing, and thus aggregation, the absorption of the excitation light is low (83% transmission) (see Fig. S7 of the Supplement 1). This shows that the aggregation process not only reduces the non-radiative decay, it also improves the absorption of the excitation light. This is an important consideration in the aggregation-induced and cross-link enhanced emission modeling.

During the annealing process, the release of water molecules leads to crosslinking between PAA polymer chains, which leads to the formation of acid anhydride as confirmed by the FTIR measurements. The crosslinking is further confirmed by the water insolubility of the strongly luminescent film. The aggregation process is further supported by TRPL measurements of non-annealed and annealed PAA films. Details of the measurements are included in Supplement 1 (see Table S1 and Fig. S2). Carrier lifetimes of 4.3, 4.7 and 3.3 ns are obtained for non-annealed, 1 minute (350 °C), 2 minute (350 °C,) annealed samples respectively. The non-annealed sample has a strong background signal in our measurement system where the excitation repetition rate is 2.5 MHz. This could be attributed to a slowly-decaying phosphorescence emission as described in detail by Zhou et al. [13]. We believe, however, that the fast decays in our measurements are from fluorescence processes. The increase in the carrier lifetime for 1 minute annealing can be attributed to suppression of the non-radiative recombination process due to the aggregation process. A decrease in carrier lifetime as we increase the annealing duration from 1 to 2 minutes is due to more efficient radiative recombination (faster radiative recombination).

Finally, both PAA and anhydrides are biocompatible products [33–35] which can be used for in vivo biomedical applications. The emission property depends sensitively on many conditions such as excitation-exposure duration and annealing conditions. This can be utilized in the development of bio-friendly sensing applications. While non-conventional organic emitters, other than PAA, are widely investigated in solution form, film-based nonconventional organic emitters could facilitate their adaptation for practical applications. In the future, ab initio molecular dynamic simulations can reveal more detailed explanation of emission mechanism on the atomic level.

5. Conclusion

In conclusion, we reveal a strong photoluminescence from a widely accessible organic poly(acrylic acid) molecule induced by a cost-effective annealing process and without complicated chemical processing. The annealing process boosts the PL intensity by a factor of more than two orders of magnitude in film-based emission. We have shown, that the emission property can be
optimized and tuned by annealing temperature, durations, solvent:PAA composition ratio in film preparation, and excitation wavelength and duration. An improved adhesion of the fluorescent film to the substrate is achieved using a porous SiC surface. The strong emission mechanism is attributed to the aggregation and crosslinking of sub-luminophores. The thermally-induced aggregation and crosslinking, and subsequent suppression of molecular motions, subdues the non-radiative relaxation of the excited carriers. These processes not only suppress the non-radiative recombination pathways, but also increase the absorption of the excitation laser by the luminescent layer. The suppression of the molecular motions is revealed directly by FTIR transmission measurements. Furthermore, the FTIR measurement reveals partial conversion of the poly(acrylic acid) into poly(acrylic anhydride). Thus, after annealing poly(acrylic acid) and poly(acrylic anhydride) coexist in the strongly luminescent films. The strong light emission from a biocompatible and non-toxic, non-conjugate polymer, which can be prepared cost effectively, can attract potential bio-medical applications such as controlled drug delivery, wavelength-conversion films and in vivo fluorescence imaging.

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Disclosures
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See Supplement 1 for supporting content.

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