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Triphenylamine-Modified Cinnamaldehyde Derivate as a Molecular Sensor for Viscosity Detection in Liquids

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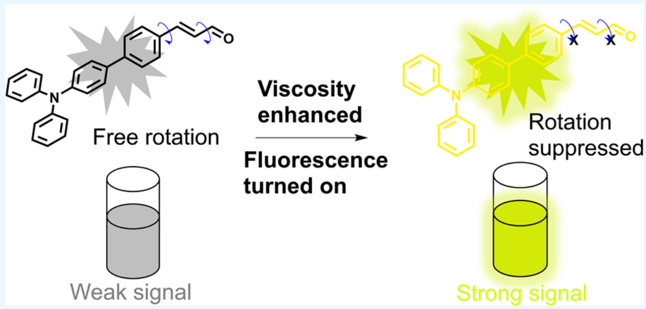
ABSTRACT: Liquid safety is considered a serious public health problem; a convenient and effective viscosity determination method has been regarded as one of the powerful means to detect liquid safety. Herein, one kind of triphenylamine-modified cinnamaldehyde-based fluorescent sensor (3-(4′-(diphenylamino)-[1,1′-biphenyl]-4-yl)acrylaldehyde (DPABA)) has been developed for sensing viscosity fluctuations in a liquid system, where a cinnamaldehyde derivative was extracted from one kind of natural plant cinnamon and acted as an acceptor, which has been combined with a triphenylamine derivate via the Suzuki coupling reaction within one facile step. Twisted intramolecular charge transfer (TICT) was observed, and the rotation could be restricted in the high-viscosity microenvironment; thus, the fluorescent signal was released at 548 nm. Featured with a larger Stokes shift (223.8 nm in water, 145.0 nm in glycerol), high adaptability, sensitivity, selectivity, and good photostability, the capability of high signal-to-noise ratio sensing was achieved. Importantly, this sensor DPABA has achieved noninvasively identifying thickening efficiency investigation, and viscosity fluctuations during the liquid deterioration program have been screened as well. We believed that this unique strategy can accelerate intelligent molecular platforms toward liquid quality and safety inspection.



■ INTRODUCTION

Fresh liquids are easily perished under ambient temperature, especially in contact with the air atmosphere, which has been regarded as one of the most common concerns all around the world.[1−3] Since the cases of health problems caused by liquid safety are growing rapidly, a convenient method for liquid safety inspection is urgent to be developed. According to studies, it is reported that various nutritious additives, including anions, cations, glucose, vitamin, etc., existed in a liquid system.[4,5] However, these nutrients accelerate the growth of mold, bacteria, and yeast, where the microenvironment is changed.[6] Typically, the viscosity can be changed during the deterioration process.[7,8] Indeed, the viscosity value is an important parameter in the liquid system, which indicates the different stages in the process of deterioration.[9,10] Although several kinds of traditional viscometers have been developed, they are only suitable for macroenvironment viscosity detection.[11−14] Notably, traditional methods need longer test times, complex pretreatment processes, and maybe destructiveness to the samples.[15−17] These shortcomings make them unsuitable in many fields, and the microenvironmental viscosity needs to be measured at a molecular level (Scheme 1).[18,19]

In recent years, photoluminescent materials have become functional optical tools for visualized detection of physical environment changes,[20−22] small-molecule monitoring,[23,24] biomarker imaging,[25] and even solid-state devices.[26,27] Commonly, molecular probes for viscosity sensing have been widely used in biological systems, as described in Table S1. It can be seen that these fluorophores are constructed by a typical donor−π−acceptor (D−π−A) molecular structure, and a twisting intramolecular charge transfer mechanism is formed in all of the molecules. Such molecular structures can rotate freely or be limited in various physical microenvironments with different consumption pathways of excited energy.[28,29] Thus, different fluorescence intensities can be seen, and viscosity changes may be tracked.[30] Based on these theories, the natural product with a D−π−A conjugated structure can be developed as an optical tool as well. Most of the previous studies relied on complex synthesis procedures, and the natural product was seldom used. With sustainable, reproducible, and degradable features, a reproducible resource has become one of the research topics in recent years, and low carbon sustainable development philosophy needs to be practiced.[31,32] As we know, a clean and reproducible resource is critical to maintaining sustainable development, and the natural prod-

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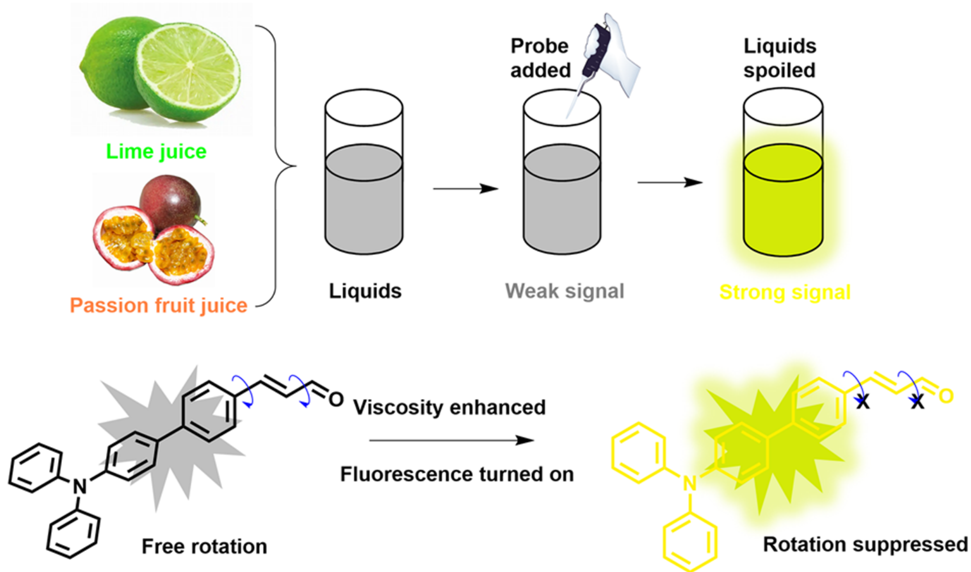
Scheme 1. Sensor Structure of 3-(4′-(Diphenylamino)-[1,1′-biphenyl]-4-yl)acrylaldehyde (DPABA) and Rational Design in Viscosity Sensing of Liquids



uct-based fluorescent sensor is regarded as a promising pathway in the construction of sensing tools.

In this work, a novel molecular sensor DPABA based on one kind of cinnamaldehyde-extracted product was established as one kind of molecular tool. To enhance the electron−donor capability, triphenylamine has been applied to act as a donor (D) and cinnamaldehyde has been used as an acceptor (A), which is extracted from the natural product cinnamon, and a flexible D−π−A chemical structure was formed. DPABA would undergo an intramolecular D−A twisting in the flexible conjugated structure, and the TICT mechanism may be used to sense the viscosity change.[33] The sensor DPABA can respond to the enhanced viscosity in a turn-on mode. The rotatable conjugated structure can act as the recognition of viscosity, which undergoes a TICT mechanism.[34,35] Experiments demonstrate that DPABA owns high sensitivity, selectivity, and photostability and can be used to investigate the viscosity variations in many commercial liquids. Furthermore, DPABA was successfully employed to evaluate viscosity fluctuations with the addition of various thickeners. Motivated by the optical properties, DPABA also can precisely track the deterioration extent by monitoring viscosity variations in the liquid microenvironment through the fluorescent technique.

■ MATERIALS AND METHODS

Chemicals and Apparatus. Detailed chemical reagents and instruments have been listed in the Supporting Information. All of the reagents have been used directly without any further purification process, and the water used in this study was triple distilled.

Synthesis Procedure of 3-(4′-(Diphenylamino)-[1,1′-biphenyl]-4-yl)acrylaldehyde (DPABA). Both 3-(4-bromophenyl)acrylaldehyde (210.0 mg, 1.0 mmol) and 4-bromo-*N*,*N*-diphenylaniline (324.2 mg, 1.0 mmol) were dissolved in toluene (10 mL), and the mixture was stirred at room temperature for over 1 h in the presence of a potassium carbonate solution (2 M). Then, the solution was refluxed overnight with an inert atmosphere of nitrogen. During the reflux procedure, 253.9 mg (1.0 mmol) of 4,4,4′,4′,5,5,5′,5′-octamethyl-2,2′-bi(1,3,2-dioxaborolane) was added dropwise. After cooling to the ambient temperature, the solvent was removed using a rotary evaporator. After purifying with DCM/petroleum ether (v/v = 2:1), the final molecular sensor DPABA was obtained (311.4 mg, 83%). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.71 (d, J = 7.7 Hz, 1H), 7.67−7.57 (m, 4H), 7.53−7.46 (m, 3H), 7.27 (dd, J = 13.8, 6.0 Hz, 4H), 7.13 (dd, J = 8.3, 2.0 Hz, 6H), 7.05 (t, J = 7.3 Hz, 2H), 6.74 (dd, J = 15.9, 7.7 Hz, 1H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 193.56, 152.32, 148.10, 147.46, 143.53, 133.20, 132.42, 129.39, 129.08, 127.71, 127.03, 124.79, 123.37. MS (ESI): m/z 376.19029 [M + H]$^+$, calcd for $C_{27}H_{21}NO$ 375.16231 [M].

Measurements of Optical Properties. The stock solution of molecular sensor DPABA was prepared in a concentration of 1 mM and stored under a lower temperature before the test. In the viscosity sensing test, various volume percentages of glycerol ranging from 0 to 99% were added into the water, and optical spectra were obtained. In the polarity test, DPABA was added to various common solvents with different polarities, and the optical spectra were operated. In the specificity test, various potential interfering analytes (100 μM) were dropped into the distilled water, and a complex solvent microenvironment was established. In a typical optical test, DPABA was then added into a quartz cuvette for investigating the anti-interference characteristic. Glycerol was stored under different temperatures (25, 5, and 37 °C) to test the temperature effect on viscosity; the solutions of DPABA were shaken uniformly before the spectra were recorded. In all measurements, the excitation wavelength was 380 nm.

Viscosity Tracking in the Spoilage Process. Lime juice and passion fruit liquids were stored at different temperatures for 1 week, and the spectra were recorded on day 0, day 2, day 5, and day 7, respectively. The relationship among the fluorescence intensity and viscosity variation was defined as below: $(\eta_n - \eta_0)/\eta_0 \sim (F_n - F_0)/F_0$, where η_n and F_n are viscosity and fluorescence intensity at day n ($0 < n < 8$), and η_0 and F_0 were the viscosity value and fluorescence intensity at day 0 and day n ($0 < n < 8$).

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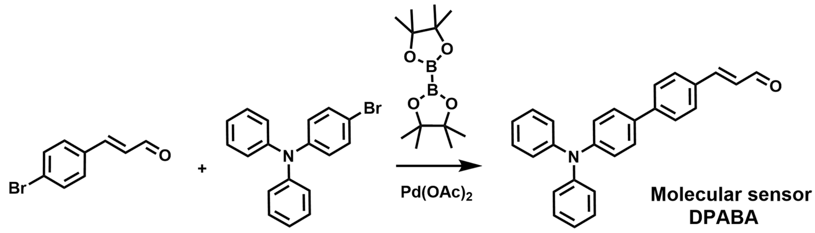

Figure 1. Synthesis of sensor DPABA.

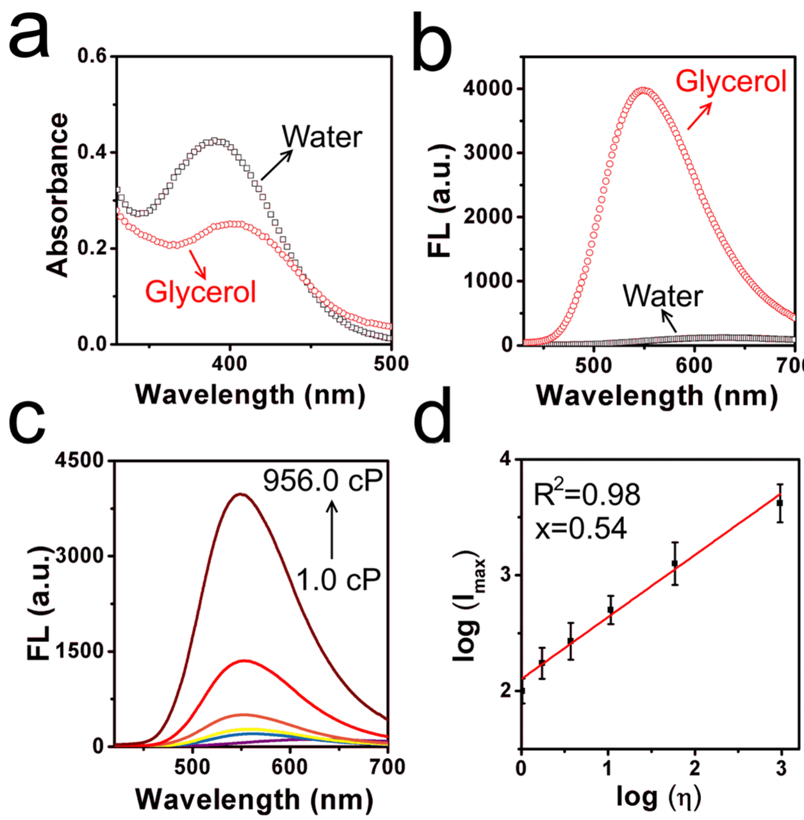

Figure 2. (a) Absorption spectra in the distilled water and glycerol. (b) Fluorescence spectra in the distilled water and glycerol. (c) Emission spectra of DPABA in the different volume fractions (f_g) of glycerol with 0−99%. (d) Fitting line among $\log I_{max}$ and $\log \eta$. λ_{ex} = 380 nm.

Theoretical Calculations. The Gaussian 09 program was utilized to perform the theoretical calculations, and the time-dependent density functional theory (TD-DFT) in the B3LYP/6-31G(d) level was applied.

RESULTS AND DISCUSSION

Molecular Sensor Design and Synthesis. The photoluminescent behavior for viscosity monitoring may be changed by the molecular charge transfer process. The donor−π−acceptor structure usually occurs in the molecular configuration, and we expect that a stronger fluorescence signal can be released in high-viscosity media. Therefore, in the designing of DPABA, one kind of natural product extracted from cinnamaldehyde was introduced to extend the conjugation, and D and A were hosted in one molecular sensor by coincidence. Thus, the TICT structure was formed. Larger conjugation of flexible molecular structures would be better for fluorescence performance to avoid autofluorescence. We hypothesized that the geometry of the sensor makes it possible to rotate freely in a low-viscosity microenvironment, and rotation can be restrained in the high-viscosity media; the conversion between the electron donor (TPA moiety) and electron acceptor (aldehyde group) may afford the inspection mechanism. Although in normal commercial liquids, the rotation is free, rapid consumption of excited energy may lead to weaker fluorescence. In spoiled liquids, the rotation is restricted, and the radiative consumption pathway will be restored. Combined with elements, DPABA was prepared through the Suzuki coupling reaction procedure within one facile step. The chemical structure is outlined in Figure 1, and the corresponding structure of DPABA was defined by ^{1}H NMR, ^{13}C NMR, and high-resolution mass spectroscopy (HR-MS), as shown in Figures S1−S3.

Optical Properties toward Viscosity. The above design strategy intrigued us to investigate the optical response capability for viscosity. Initially, the spectra in the water and glycerol were evaluated. As shown in Figure 2a, the absorption peak in the high-viscosity glycerol was red-shifted from 391.4 to 403.0 nm when contrasted to that in the lower-viscosity pure water.

This red-shifted data may be ascribed to the prevented rotation in the high-viscosity environment, and the sensor

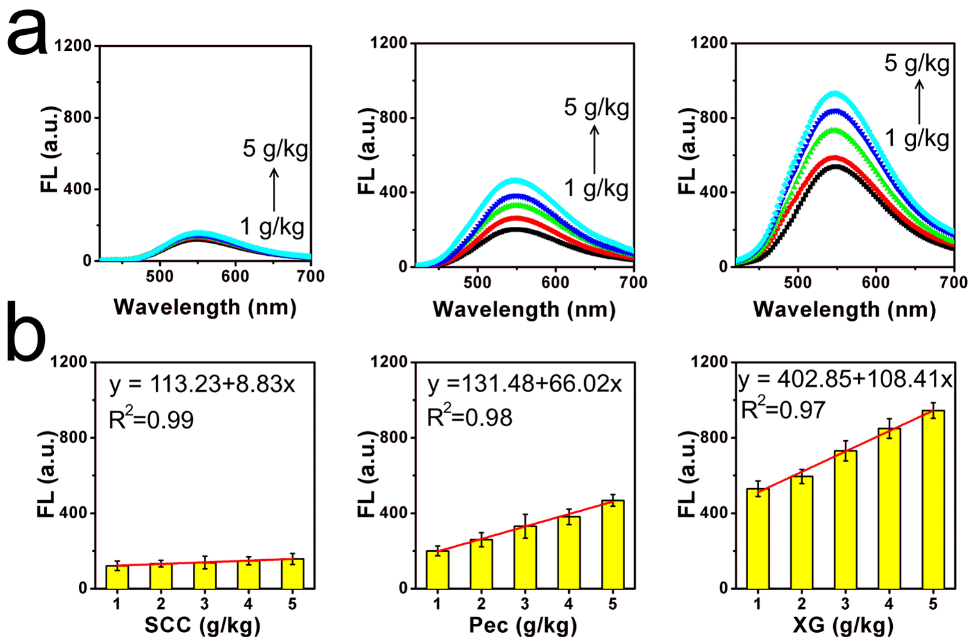

Figure 3. (a) Emission spectra in the solutions with various mass amounts of SCC, Pec, and XG from 1 to 5 g/kg. (b) Fluorescence intensities in the solutions with various mass amounts of SCC, Pec, and XG and corresponding fitting lines. λ_{ex} = 380 nm.

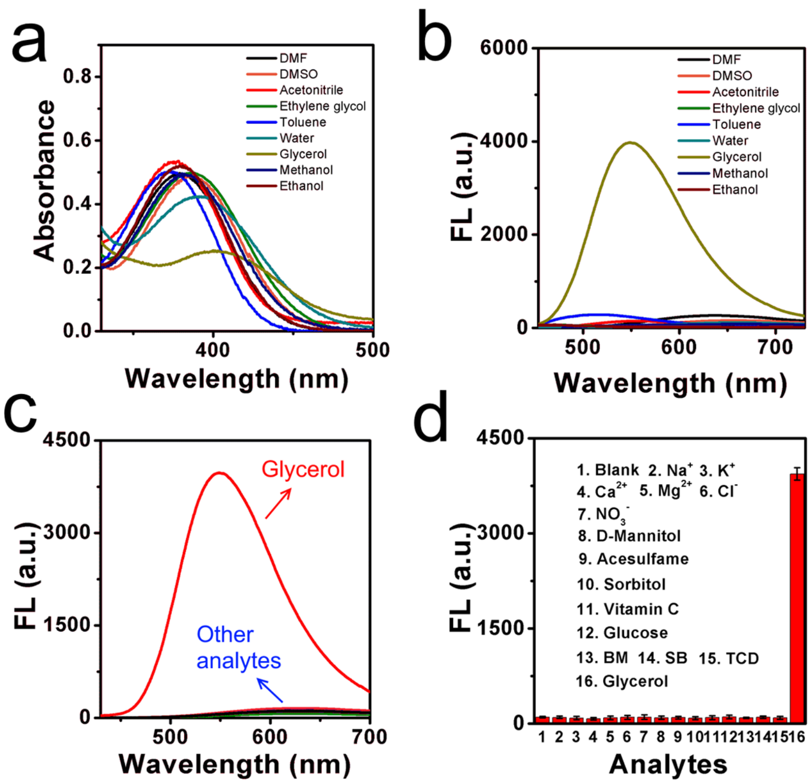

Figure 4. (a) Absorption spectra of the sensor DPABA in different solvents. (b) Emission spectra of the sensor DPABA in different solvents. (c) Emission spectra of DPABA for related analytes, 1. blank, 2. Na^+, 3. K^+, 4. Ca^{2+}, 5. Mg^{2+}, 6. Cl^-, 7. NO_3^-, 8. D-mannitol, 9. acesulfame, 10. sorbitol, 11. vitamin C, 12. glucose, 13. beet molasses (BM), 14. sodium benzoate (SB), 15. trisodium citrate dehydrates (TCD), and 16. glycerol. (d) Fluorescence intensity responses of DPABA for various analytes under an excitation of 380 nm.

molecules would be parallel stacked.[36,37] On the other hand, considerable variation in the emission spectra was noted, and a 40-fold enhancement was found, as shown in Figure 2b. Moreover, fluorescence response in different viscosity media was investigated with the gradually increasing proportions of glycerol in a water–glycerol mixture. A linear relationship can be established by fitting the Förster–Hoffmann equation (R^2 = 0.98, x = 0.54), as shown in Figure 2c,d.[38,39] Furthermore, the fluorescence lifetime τ of sensor DPABA also fits well with the solvent viscosity changes (in Figure S4); the results further confirmed the fluorescence enhancement mechanism. The detection limit was determined as 1.253 cP, as shown in Figure

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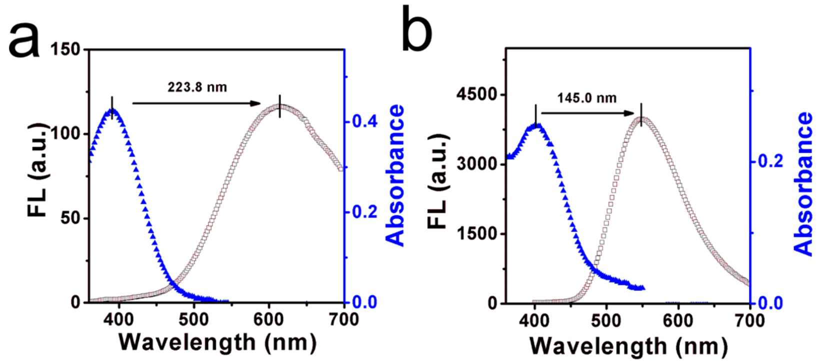

Figure 5. (a) Fluorescence response and absorption spectra of DPABA in water. (b) Fluorescence response and absorption spectra of DPABA in high-viscosity glycerol.

S5. In addition, as shown in Figure S6, when glycerol was stored under different temperatures, DPABA released a higher fluorescent signal when stored under 5°, while the signal became weaker with the enhancement of temperature. To develop the potential application value of sensor DPABA, we tested the application of sensor DPABA in various viscosities of liquids. As shown in Figure S7, 11 kinds of commercial liquids were selected herein. DPABA released various fluorescence signals in these liquids, which indicated that DPABA could be instrumental in viscosity detection as a molecular tool. Detailed fluorescent intensities in these liquids are collected in Table S2. In addition, the viscosity values have been measured by a viscometer as well, as shown in Table S3. The experimental data were consistent with the calculated data obtained by the fluorescent method. Afterward, many kinds of thickeners have been used to enhance the homogeneity, consistency, and stability of the liquid system.[40] Thus, we selected sodium carboxymethyl cellulose (SCC), pectin (Pec), and xanthan gum (XG) as the representative species, and different viscous media have been built with various weight amounts of thickeners. With the additional amount of food thickeners, a stronger signal was released in the SCC, Pec, and XG solutions, which may be attributed to the viscosity enhancement caused by these additives (in Figure 3a). In Figure 3b, the fitting lines have been established, and different linear slopes can be found. A large linear slope value (108.41) was observed in the XG solutions, while a lower linear slope value (8.83) occurred in the SCC solutions. Overall, optical studies indicate that DPABA is suitable to be an intelligent molecular tool for viscosity investigation.

Adaptability, Selectivity, and Photostability Investigation. Next, the fluorescence spectra changes of sensor DPABA to the polarity was investigated. Herein, several solvents with different polarities were chosen, including dimethylformamide (DMF), dimethyl sulfoxide (DMSO), acetonitrile, toluene, water, glycerol, methanol, and ethanol. In contrast to the absorption peak in other common solvents around 380 nm, the peak was red-shifted toward 401 nm in glycerol, as shown in Figure 4a. High-viscosity glycerol may lead to the parallel stacking of sensor DPABA, and the conjugation can be enlarged.[41] A solvent dependence was observed. In addition, in contrast to the stronger signal that occurred in the glycerol system, weaker signals were released in the other common solvents (Figure 4b). The results indicate that DPABA is inert to the polarity changes, and the potential adaptability of DPABA in most common liquids has been confirmed. Detailed absorption and emission intensities in these solvents have been shown in Table S4. To understand the selectivity of DPABA in a complex liquid system, various relevant liquid species, including the metal ions, anions, and common food additives (including the D-mannitol, acesulfame, sorbitol, VC, glucose, BM, SB, TCD) were added individually into the sensor DPABA solutions, and the fluorescence responsibility was measured. In Figure 4c, a dramatically enhanced fluorescence emission at 548.0 nm was triggered by the high-viscosity microenvironment in glycerol, while the introduction of other species caused negligible fluorescence changes even at a high concentration (100 μM). A corresponding fluorescence intensity histogram is shown in Figure 4d. This result indicates that DPABA is highly sensitive to viscosity and cannot be interfered with the fluorescence signal release. Given that a large Stokes shift may lead to the enhancement of the signal-to-noise ratio,[42] the Stokes shift was investigated as well. As shown in Figure 5a,b, it can be found that the Stokes shift was 223.8 nm in water and 145.0 nm in glycerol, respectively. Such a large Stokes shift might enable DPABA to detect the viscosity without autofluorescence. Therefore, sensor DPABA was able to discriminate viscosity changes from other potential interferences.

Given that pH changes in liquid samples may cause fluctuations in the fluorescence intensity, the pH effect on the fluorescence response of DPABA was investigated. Subsequently, the novel molecular sensor is essentially pH-insensitive over a range of 2.0−9.0, suggesting its feasibility of applications in a liquid microenvironment. In Figure S8, the sensor DPABA maintained a constant weak emission signal across the pH range, which indicates the potential application in complex commercial liquids. Inspired by these results, corresponding photostability in multiple liquids was investigated; the sensor DPABA was added to 11 kinds of liquids, including water, lime juice, peach tea, etc., and the test was performed under continuous irradiation within 1 h. More than 98% fluorescence intensity still existed during the test time range, demonstrating the excellent photostability of sensor DPABA and confirming its potential applicability in these common liquids, as shown in Figure S9. Moreover, the stability of sensor DPABA in various solutions for 7 days was investigated as well. The fluorescence emission intensities were recorded at day 0, day 2, day 5, and day 7, which were found to be maintained stable during the storage time, as shown in Figure S10.

With the experimental results, the theoretical calculations of the energy gap and oscillator strength f_{em} were performed using Gaussian 09 software, as shown in Figure S11. The HOMOs

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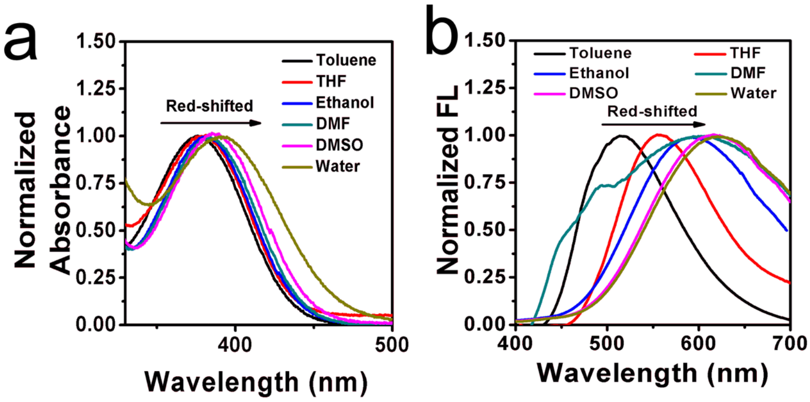

Figure 6. (a) Normalized emission spectra of DPABA in toluene, tetrahydrofuran (THF), ethanol, DMF, DMSO, and water. (b) Normalized absorption spectra of DPABA in different solvents.

were mainly located in the triphenylamine group and the LUMOs were found over the conjugated aldehyde group. A strong charge transfer between the triphenylamine group and conjugated aldehyde group and an ICT process occurred. The energy gap at 0° was calculated to be 2.9966 eV, while the energy gap at 90° was calculated to be 3.0701 eV. Meanwhile, corresponding f_{em} values were 0.7545 (0°) and 0.0008 (90°), respectively. A typical twisted excited state can be utilized to determine the microenvironment viscosity changes with the intramolecular rotation. Moreover, the solvatochromism of DPABA was measured as well. A typical red-shifting phenomenon was found in both spectra, as shown in Figure 6a,b. This phenomenon further confirmed the stabilization of ICT occurring among the triphenylamine and conjugated aldehyde groups.

Deterioration Process Monitoring. Encouraged by optical spectra results, we further tested the DPABA's capability in tracking the liquid deterioration process. Thus, the lime juice and passion fruit juice were stored under different temperatures (ambient temperature and fresh-maintenance temperature) for 1 week, respectively. At first, the deterioration processes were recorded by a camera, as shown in Figure 7a. At the beginning of a couple of days, transparent and clear appearances were found in these two liquids. In the next few days (especially after day 5), by contrast, the floating objects occurred and turbid phenomena could be observed in both liquids. On the contrary, a normal appearance was still maintained even though the storage time was extended to 5 days when under the lower storage temperature, as shown in Figure S12a. Moreover, the deterioration process was measured through the fluorescent method as well (in Figures 7b and S12b). Around 11.8 and 14.8% increases were found in the fluorescence spectra of lime and passion fruit juice when the storage time reached 7 days under ambient temperature. However, the fluorescence signal increased in a limited range under lower storage temperature, with only 6.3 and 6.8% enhancement in lime juice and passion fruit juice, respectively. These solid images and spectra data demonstrated that sensor DPABA can be applied to sensitively detect the microenvironment viscosity fluctuations via the fluorescent technique.

As stated above, the viscosity values have been determined by a viscometer as well. When these two kinds of liquids were stored under ambient temperature, the viscosity values were enhanced by 22.5 and 25.1%, respectively. By contrast, the viscosities of these two liquids enhanced only by 10.1 and

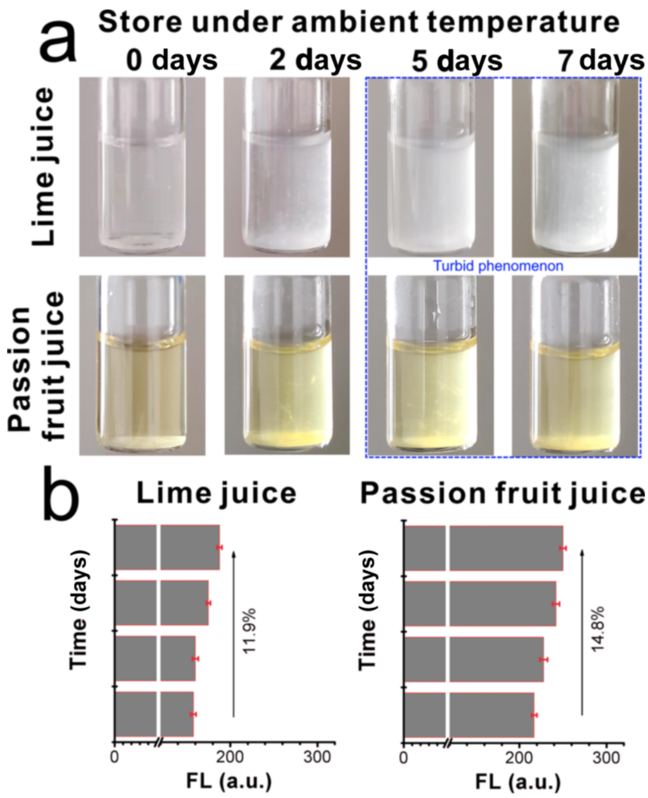

Figure 7. (a) Digital images of the lime juice and passion fruit juice that were stored under ambient temperature for 7 days. (b) Emission intensity of lime juice and passion fruit juice when stored under ambient temperature for 7 days. λ_{ex} = 380 nm.

12.1% when stored at the lower storage temperature (in Figure 8a,b). The results were consistent with the fluorescent investigation results. Notably, a fitting linear relationship was established between the viscosity increase and the fluorescence intensity enhancement, as shown in Figure 8c. These results not only verified that sensor DPABA is suitable to visualize the microenvironmental viscosity variations but also offered a possible and convenient pathway to track the deterioration process through the fluorescent technique.

In addition, *Escherichia coli* was one of the representative bacteria that occurred in spoiled liquids, which destroys the nutritional components seriously, and the corruption will be accelerated. These kinds of bacteria are considered an indicator

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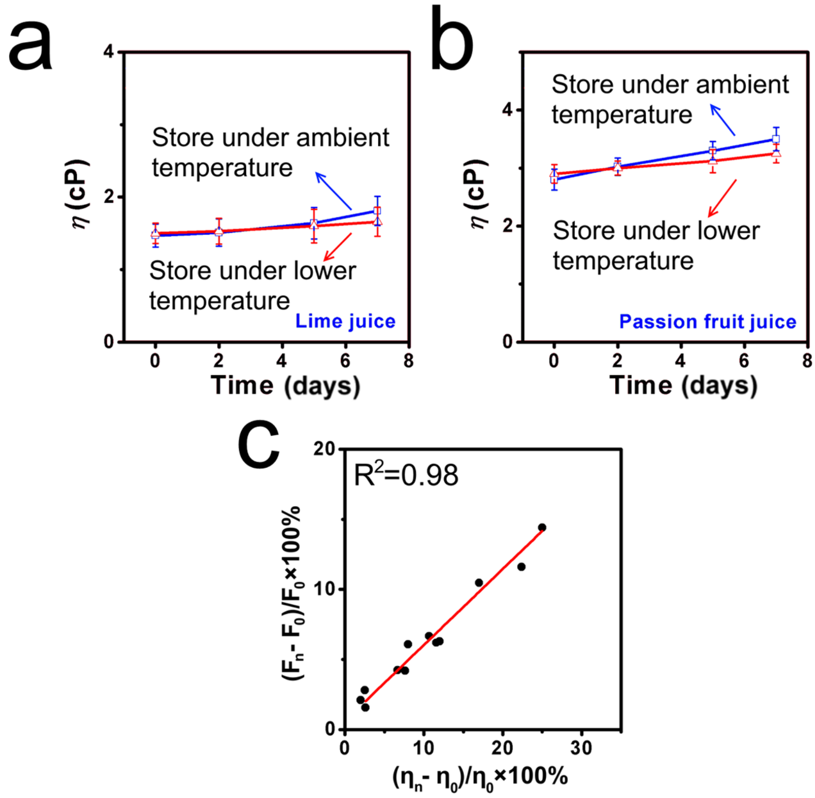

Figure 8. (a) Viscosity values of lime juice when stored under ambient temperature within 7 days. (b) Viscosity values of passion fruit juice when stored under lower temperature within 7 days. (c) Fitting line among the fluorescence intensities and viscosity values.

of deterioration, which can cause diarrhea and malnutrition, or even death. Thus, an additional experiment containing *E. Coli* was performed in the presence of sensor DPABA in these liquids. As shown in Figure S13a, several kinds of floating objects and precipitates can be found when the lime juice and passion fruit juice are stored under a lower temperature for 3 days. The fluorescence intensity increased 5.0 and 6.1%, respectively. Corresponding viscosity values increased 6.2 and 8.7%, respectively. In comparison, the situation became even worse when stored under ambient temperature, as shown in Figure S13b. The increase in fluorescence intensity was 9.8 and 12.8%, and the corresponding viscosity value increased 16.7 and 20.8%, respectively. A serious deterioration phenomenon was observed. From the results, it can be found that higher fluorescence intensity increase and viscosity enhancement occurred after the addition of *E. Coli*, and the deterioration speed was accelerated. Thereby, these results further collectively manifested the effectiveness of sensor DPABA.

In this report, cinnamaldehyde-based natural product DPABA has been designed, and a triphenylamine derivate donor and conjugated aldehyde acceptor were synthesized successfully depending on the restriction of a rotatable conjugate structure and the TICT effect. DPABA possessed a typical large Stokes shift in both water and glycerol systems and high photostability in via one facile step. Sensor DPABA displayed a turn-on mode toward viscosity sensing in various commercial liquids, where high selectivity in the presence of related additives and a high sensitivity coefficient (x = 0.54) were observed. Furthermore, for a better understanding of the liquid spoiled process, this natural product-based sensor DPABA was utilized to quantitatively monitor viscosity changes in a noninvasive and in situ pathway. We expected that this strategy can intersect the original intention of sustainable molecular design with the fluorescent imaging technique, which will accelerate the perfection of a multifunctional molecular platform.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.3c00488.

Fluorescence analysis; Förster−Hoffmann equation; ^{1}H NMR spectra; ^{13}C NMR spectra; mass spectra; detection limit; fluorescence spectra under different temperatures; fluorescence spectra in nine kinds of liquids; photostability under different pH values; photostability analysis in nine kinds of beverages; and theoretical calculations (PDF)

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Author Contributions

This manuscript was written through the contributions of all authors. All authors have approved the final version of the manuscript.

Notes

The authors declare no competing financial interest.

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