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Optimized erucic acid-based extract as a natural probe for viscosity detection of liquid safety

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ABSTRACT

As the main substate participating during the liquid metamorphism processes, the abnormal micro-environmental viscosity is closely associated with the occurrence and development of objective circumstance degradation. Thus, it is of great importance to establish the convenient detection method toward viscosity monitoring. This work is focused on extracting one kind of erucic acid-based molecular probe (EAd) for sensing viscosity fluctuations in liquids. The electron donor phenolic hydroxyl group and acceptor carboxyl have established in this molecular structure *via* a coincidental pathway. Twisted intramolecular charge transfer (TICT) was formed, and the rotatable conjugate structure was utilized as the recognition site. Due to the restriction of rotatable parts, higher emission fluorescent signal can be found in the high-viscosity micro-environment. And a large Stokes shift (83.3 nm in glycerol), narrower energy band, high selectivity (x = 0.56), adaptability, sensitivity, and good photo-stability, the applications of signal releasing in the complex liquid system can be achieved. Moreover, this natural probe EAd can identify the thickening efficiency in a non-invasively mode, in particular, viscosity fluctuations have been screened and spoiled samples can be distinguished by analyzing signal intensities. We hope that natural scaffold will provide a new reference toward liquid quality and safety inspection.

1. Introduction

Micro-environmental viscosity serves as an important physical parameter in the liquid system, while the abnormal fluctuations of viscosity are closely related with the met-amorphism process (Hajikhani and Lin, 2022; Zhang et al., 2024; Zhou et al., 2023). Commonly, various nutritious additives, such as cations, anions, small-molecular-weight glucose, vitamin, etc., displayed key roles in maintaining homeostasis in liquids (Cox et al., 2021; Moradi et al., 2022). Especially, the growth of bacteria, mould and yeast depends on the aforementioned nutrients, which can cause obvious viscosity changes during the deterioration process (He et al., 2020). As a vital important marker of the micro-environment, the viscosity should be kept in normal state to maintain the micro-environmental physiological activities (Li et al., 2024; Ma et al., 2020). Obvious differences of the viscosities is associated with the development stages of deterioration (Du et al., 2022; Gao et al., 2021; Li et al., 2024). Therefore, monitoring the slight viscosity changes in a molecular way is important to better understand the deterioration functions and distinguish the spoiled ones from the fresh liquids. To date, many kinds of viscometer have been developed toward viscosity determination (Lee et al., 2020; Mäkelä et al., 2020; Sagdeev et al., 2020). These methods still suffer from several shortcomings, such as complex pre-treatment processes, longer test time, destructiveness to the sample and heavily rely on the device (Pan et al., 2023; Wu et al., 2023; Yang et al., 2022a). Compared to the traditional methods, the microscopic viscosity to be detected at a molecular level is essential, which can break through the shortcomings limit of macroscopic detection methods (Yang et al., 2022b; Ye et al., 2021).

In recent years, luminescent materials have been developed and become a powerful tool for viscosity tracing in liquid systems due to the advantages of high sensitivity, fast response, noninvasive detection, and *in situ* sensing (Chen et al., 2020; Liu et al., 2022; Xu et al., 2022; Liu et al., 2023; Wu et al., 2023; Xu et al., 2023). To date, various fluorescent probes have been developed and been utilized to detect the

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viscosity in the biological field, as shown in Table S1 (in the Supplementary Material). It can be found that most of previous studies are relying on the multifarious synthesis process, large amount of organic solvents consuming, time wasting, etc. On the contrary, natural products have a variety of highly ordered hierarchical structures, and multiple molecules can offer the unique functionalities such as physical, chemical, or biological stimuli, which can provide a potential means for viscosity detection. Thus, complex synthesis procedures and the consumption of multiple toxic organic solvents can be avoided. Efficient utilization of clean and reproducible resource is critical to maintaining sustainable development, natural product-based fluorescent sensor is regarded as one kind of promising sustainable energy source in the construction of multi-functional molecular platform (Hou et al., 2020; Sabzehmeidani and Kazemzad, 2022). It is worth to mention that the typical do-nor-π-acceptor (D-π-A) molecular structure can formed into a twisted intramolecular charge transfer (TICT) effect (Song et al., 2022). Such kind of molecules can rotate freely in low-viscous solutions, and the relaxation process of electronically excited dyes can be carried out via non-radiative transitions (Miao et al., 2022). While in high-viscous media, the intramolecular motions are inhibited, and energy is released in radiative decay paths, stronger fluorescence can be found (Li et al., 2024b). Therefore, it remains urgent and important to explore the structure–activity relationships and to develop several kinds of certain natural products for sensor tool.

Natural rosemary is a well-known herb of Lamiaceae family originated from Mediterranean region, which has been utilized as a spice of folk medicine for a long time. It has been revealed that rosemary owns many physiological active ingredients, such as carnosic acid, carnosol, erucic acid, *et al.*, which are suggested to contribute to the anti-inflammatory effects and glucose-lowering functions. Based on the main active component and easily obtained raw materials, herein, one kind of natural product erucic acid (EAd) extracted from the rosemary plant was established as one kind of molecular probe. Based on the inherent electron donor phenolic hydroxyl group (D) and electron acceptor carboxyl group (A) hosted together, a flexible D-π-A chemical structure was formed, the intramolecular charge transfer (ICT) effect was shaped. We hypothesized that EAd would be able to undergo intramolecular D–A twisting phenomenon and may hold great promise in viscosity sensing (Wang et al., 2020; Xiao et al., 2021). In the emission spectra test, EAd displayed good viscosity-responsive property with the viscosity sensitivity coefficient x = 0.56. It displayed 52-fold signal enhancement with the increase of viscosity at maximum. Adequate selectivity, adaptability, and photo-stability endowing the natural probe EAd with the capacity commercial utilization. The capacity of evaluating thickening efficiency was confirmed. A DFT calculation was performed to demonstrate the sensing mechanism, a TICT program from the donor to the acceptor was executed (Liu et al., 2024; Sun et al., 2021). It was expected that the natural probe EAd can precisely track the deterioration process by monitoring viscosity variations at a molecular level. Overall, we believe that our work will promote the development of the natural product as a luminescent tool (platform) for liquid safety inspection *via* a green and sustainable way.

2. Experimental section

2.1. Materials and apparatus

The chemical reagents in this study were directly obtained from Macklin Biotechnol-ogy (Shanghai) Co., Ltd, and Shanghai Aladdin Bio-Chem Technology Co., Ltd. All the reagents and solvents were used without any further purification. Milli-Q water purifica-tion system (Millipore, Bedford, MA, USA) was used to prepared the deionized (DI) water. Nuclear magnetic resonance (NMR) spectra were obtained with Bruker AVANCE III HD 600 NMR Spectrometer, with TMS internal standard in DMSO-d_6. High resolution mass spectra (HR-MS) were performed through Aglient 7250 & JEOL-JMS-T100LP AccuTOF mass spectrometer. Fluorescence spectra were recorded by a Hitachi F-7000 fluorescence spectrophotometer. Absorption spectra were detected on a Hitachi U-3010 UV–Vis spectrophotometer. The viscosity determination test was performed on a rotating viscometer (DV2T, Brookfield, AMETEK Corp., USA). The reagents and instruments were described in the Supplementary Material.

2.2. Preparation of plant extract erucic acid (EAd)

Natural rosemary was freeze-dried as the powder, then soaked into 80 % ethanol with continuous shaking at 80 °C for 3 h. The solution was led to filtration, ethanol extract was concentrated under vacuum through the rotary evaporator. The obtained product was dissolved in distilled water and partitioned with n-hexane. Controllable fractions of n-hexane and water mixture were utilized, and obtained organic solution was evaporated under vacuum. Afterwards, the obtained extract was purified by flash column chromatography over silica gel to give the erucic acid (EAd) as a pale yellow solid, yield 1.1 %. ^{1}H NMR (600 MHz, DMSO) δ 12.13 (s, 1H), 8.90 (s, 1H), 7.51 (d, J=15.9 Hz, 1H), 7.00 (s, 2H), 6.43 (d, J=15.9 Hz, 1H), 3.81 (s, 6H). ^{13}C NMR (101 MHz, DMSO-d_6) δ 168.42, 148.53, 146.52, 138.57, 125.09, 116.55, 106.59, 56.57. HRMS (ESI): m/z calculated for 224.21200, found: 225.21204 $[M+H]^+$.

2.3. Measurements of optical properties

The molecular probe EAd was dissolved in DMSO to prepare the stock solution with the concentration of 1 mM, and stored under the temperature of 5 °C before the test. EAd solutions (10 μM) was added into different volume fractions of distilled water and glycerol mixtures (from 0 % to 99 %), and each sample were recorded by the fluorescence spectra and UV–Vis. To prepare different polarities of liquid system, the common solvents including the EtOAc, methanol, THF, toluene, etc. were selected to test the emission properties. In the specificity test, the solutions with various potential relevant analytes (including NaCl, K_2CO_3, VC, sodium benzoate (SB), etc., 50 μM) were prepared with distilled water, and EAd solutions were added as well. The fluorescence emission spectra were obtained by the fluorescence spectrometer. In the temperature effect experiment, EAd was diluted with the glycerol under different temperatures, including the normal body temperature (37 °C), fresh-maintenance temperature (5 °C) and ambient temperature (25 °C). In all measurements, the mixed solutions were transferred to the quartz cell. The excitation wavelength was set as 320 nm.

2.4. Viscosity tracking in the spoilage process

The raspberry fruit juice and lemon liquids were brought from the local supermarket, and flesh floats and sediments were removed. Before the storage process, the bubbles were removed as well, and shaked thoroughly with the transparent glass bottle. Then, both fresh liquids were stored at different temperatures (25 °C, 5 °C & 37 °C) when exposed to the air for 7 days. During the storage process, the fluorescence emission spectra were recorded within this time interval (at day 0, day 2, day 4, and day 7) with the addition of molecular probe EAd dilution (10 μM). The relationship between fluorescence signal intensity and viscosity value has been established: $(F_n\text{-}F_0)/F_0 \sim (\eta_n\text{-}\eta_0)/\eta_0$, F_n & F_0 fluorescence intensity at day n and day 0, η_n & η_0, viscosity value at day n and day 0.

2.5. Theoretical calculations

The calculations were performed using the Gaussian 09 program, ground state structure of the rotor was optimized using time-dependent density functional theory (TD-DFT) by using the B3LYP/6-31G(d) level of theory. The energy gap and oscillator strength fem were calculated as well.

3. Results and discussion

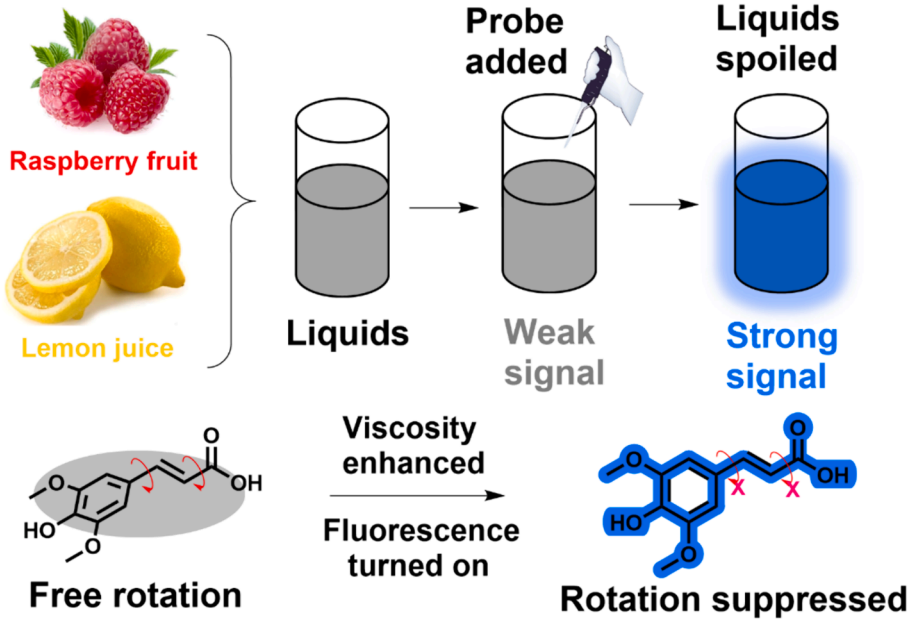
3.1. Molecular probe selected and extracted strategy

Several kinds of natural products can be used as molecular tools for physical micro-environmental viscosity sensing, the ingenious selection of a powerful optical probe is highly desirable. The natural extract EAd was found to bear phenolic hydroxyl and carboxyl groups together with the conjugate single and double bond, which exhibited a flexible and rotatable chemical structure. Through the linkage with D and A, this kind of natural probe might display different emission fluorescence and sensing properties toward viscosity *via* the potential rotation of conjugated bond. In the low viscosity media, weaker fluorescence emitted, while in the higher viscosity liquid, stronger signal came out, a turn-on mode can be found. We hypothesized that the signal change phenomenon was attributed to the restriction of rotatable part, in normal commercial liquids, the rotation is free, rapid consumption of energy may lead to weaker fluorescence, while in spoiled liquids, the rotation is restricted, and the energy consumption by the non-radiative pathway will be reduced, and the transition of TICT and ICT was activated, detailed chemical structure and sensing mechanism was shown in Scheme 1. Thus, the capability of viscosity sensing was existed objectively. In detail, the molecular probe EAd was extracted from the rosemary and has been purified through the silica gel, which was utilized directly. The chemical structure and detailed relative molecular mass of probe EAd were defined by ^{1}H NMR, ^{13}C NMR and high-resolution mass spectroscopy (HR-MS), as presented in Fig. S1-Fig. S3 (in the Supplementary Material).

3.2. Responsive properties toward viscosity

The above selected strategy intrigued us to investigate the optical responsive proper-ties toward viscosity. Initially, the fluorescence and absorption spectroscopic properties of natural probe EAd were investigated. As shown in Fig. 1a and 1b, a remarkable fluorescence signal can be observed in the high viscous glycerol, 52-fold enhancement was clearly found. Moreover, the physical properties of quantum yields and fluorescence lifetime in low viscosity water and high viscous glycerol have been collected as well, as shown in Table S2. There is nearly no fluorescence emission intensity in the water, and this remarkable difference in the intensity suggests that natural probe EAd could serve as a molecular tool for viscosity detection. Comparatively, natural probe EAd exhibited an ab-sorption peak at 329.3 nm in the high-viscosity glycerol, which was slightly red-shifted from the peak 324.9 nm in the lower viscosity pure water. The parallel stacking of molecular probe in the glycerol and the conjugation may be enlarged (Ge et al., 2023; Zhang et al., 2024). Furthermore, the viscosity-responsive properties of natural probe EAd was explored in different volume fractions of glycerol and water mixtures, as shown in Fig. 1c. With the increment of glycerol, the emission intensity increased gradually, which may be attributed to the inhibited rotation of conjugated bond in the viscous environment. Moreover, a good linear relationship (R^2 = 0.98) of the maximum emission intensity against viscosity from 1.0 cP to 956.0 cP was observed by fitting the Förster–Hoffmann equation, as shown in Fig. 1d (Wang et al., 2023). The viscosity-sensitive coefficient (x) was calculated as 0.56, and the detection limit was determined to be 1.253 cP by using the commonly equation LOD=3 σ/S, where σ means the standard deviation of a blank and S represents the slope of calibration line, as shown in Fig. S4 (Ma et al., 2024). These detailed results further confirmed natural probe EAd can potentially serve as an excellent viscosity-responsive molecular tool in a relatively large viscosity range. Besides, the viscosity sensing capability under different temperatures were investigated, since temperature is one of the key factors that can affect the viscosity. It can be found that the fluorescent intensity increased when the glycerol was stored under lower temperature, while the signal became weaker when under a higher temperature, as shown in Fig. S5. The test results further illustrate that natural probe EAd can potentially work as a sensitive molecular tool for viscosity tracking in solutions. In order to confirm the applications in commercial liquids, we tested natural probe EAd in ten kinds of liquids, including water, pomelo juice, pear juice, raspberry fruit juice, milk, lemon juice, mango juice, watermelon juice, edible oil, and glycerol, as shown in Fig. S6. Detailed fluorescent intensities in those liquids were collected in Table S3. In addition, the viscosities of these commercial liquids were measured by the viscometer as well, and the calculated results in Table S4 were inconsistent with those by the fluorescent method. We can clearly find that various fluorescence intensities in these commercial liquids, suggesting the natural probe EAd showed enormous potential to be utilized in the commercial complex samples.

The thickeners can enhance the consistency, homogeneity and



Scheme 1. Chemical structure of natural probe EAd and sensing mechanism.

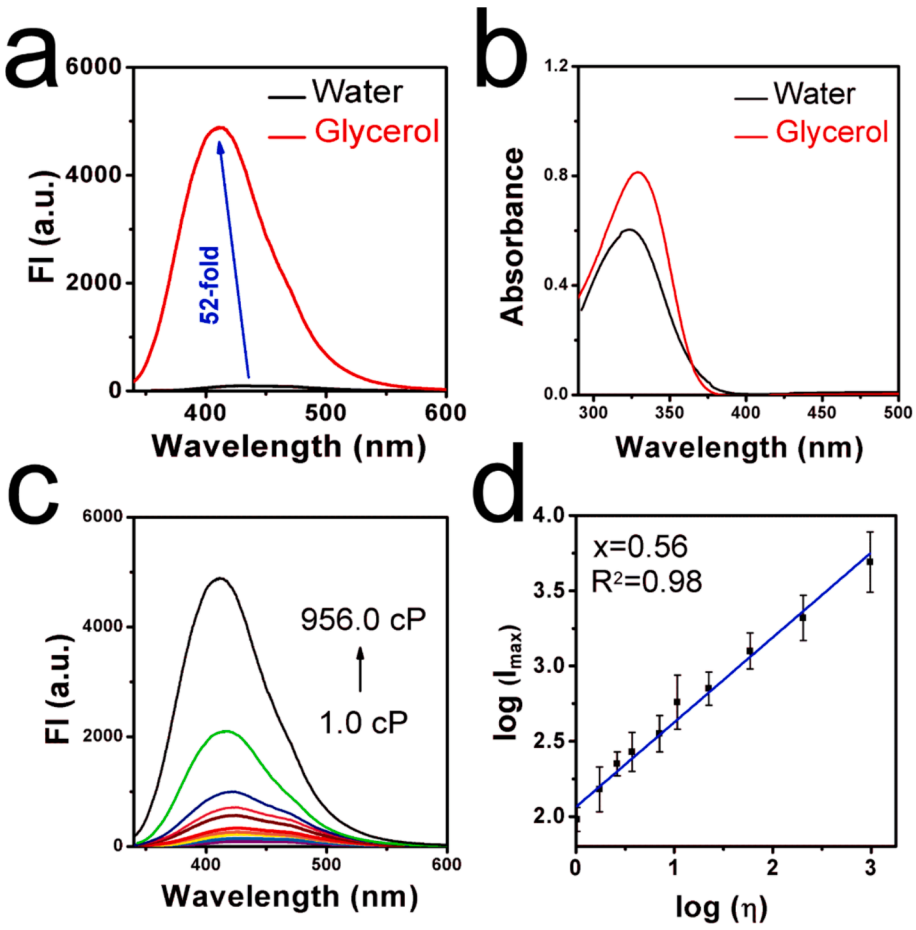


Fig. 1. (a) Fluorescence spectra of the natural probe erucic acid (EAd) (10 μM) in water and glycerol. (b) Absorption spectra of the natural probe erucic acid (EAd) (10 μM) in water and glycerol. (c) Fluorescence emission spectra of the natural probe erucic acid (EAd) (10 μM) in the water/glycerol mixture with different glycerol volume fraction of 0 %~99 %). (d) The linear relationship between log I_{max} and log η.

stability in commercial liquids, and have been one of the main components as food additives (Pirsa and Hafezi, 2023). And therefore, we further explored the sensing capability of natural probe EAd with the existence of three kinds of food thickeners, such as SCC, Pec, and XG. As shown in Fig. 2a, the fluorescence spectra were first recorded with the addition of various amounts (1.0 g/kg-5.0 g/kg) of above-mentioned thickeners. Fluorescent intensities were gradually increased with adding amounts of the food thickeners, viscous media was formed in a certain way. In Fig. 2b, it is worth to mention that a positive relationship was existed between the mass concentrations of food thickeners and fluorescence intensities. And we can find that its fluorescence in the xanthan gum (XG) reached the highest, indicating that XG exhibited the best thickening efficiency, while the fluorescence in sodium carboxymethyl cellulose (SCC) occupied the lowest, which means the thickening efficiency of SCC is lower. Therefore, we can conclude that the natural probe EAd can be appropriately acted as the intelligent fluorescent probe toward viscosity identification.

3.3. Photophysical properties investigation

Next, the photophysical properties of natural probe EAd in relation to the stability in various liquids with different polarities were investigated. Herein, the fluorescence emission and absorption spectra of EAd in eight kinds of common solvents with different dielectric constants (ε) were investigated. A remarkable fluorescence signal peak at 420 nm was found in the emission spectra of high-viscosity glycerol, as shown in Fig. 3a. And the intensities of emission spectra in other low viscous solvents were found weaker. The absorption spectra in these common solvents were distributed around 320 nm. Negligible peak wavelength can be observed, as shown in Fig. 3b. Corresponding fluorescent im-ages were exhibited in Fig. 3c, bright emission signal can be observed only in the glycerol, while weak signal was released in other liquid samples. Significant differences can be visualized from the naked-eye. High molar extinction coefficient in the water can make it possible to provide a low fluorescence background in liquid imaging. Detailed photophysical properties of the sensor EAd in these solvents were collected in Table S5. These test results indicated that the fluorescence signal cannot be affected by the solvent polarity. And the stability in various solvents was extremely well, which can be helpful to avoid the complex polar influence.

Afterward, the selectivity and anti-interference of natural probe EAd to various potential analytes in the liquid system were investigated, since the micro-environment in the liquid system is complex. With the inclusion of ions (Na^+, K^+, Ca^{2+}, SO_4^{2-}, Cl^-, NO_3^-, CO_3^{2-}), common liquid contents (gallic acid, acesulfame, sorbic acid, erythorbic acid, sodium benzoate, beet molasses, trisodium citrate dehydrate) and vitamin C, the FI response intensity of EAd was extremely lower. As shown in Fig. 3d and 3e, negligible changes can be found in the fluorescence intensity histogram with these additives. Only dramatically enhanced fluorescence signal was triggered by the glycerol, which showed that the fluorescence signal cannot be influenced by the various food additives, and FI response of EAd to viscosity (i.e., glycerol) was higher than for other species. Moreover, the photostability in different pH has been evaluated, as shown in Fig. S7, six kinds of buffer solutions in the pH range of 3.0–9.0 were prepared, with the final concentration of 10 μM. The emission intensities were found to be even lower, a negligible

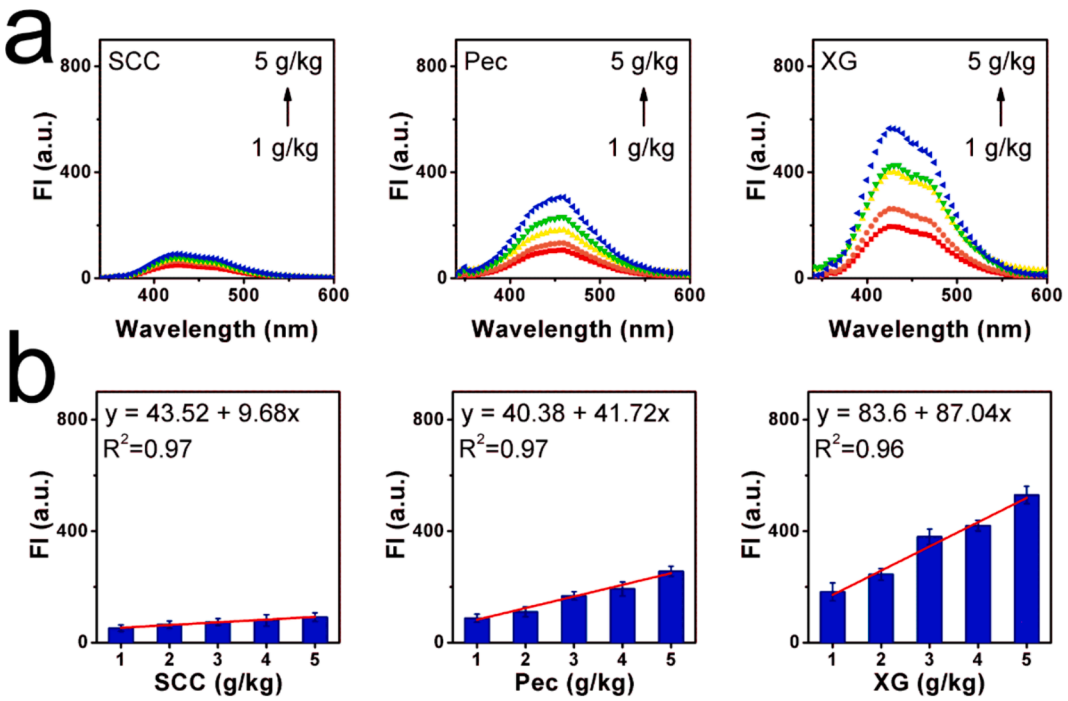


Fig. 2. (a) Emission spectra of the molecular probe EAd in the SCC, Pec, and XG solutions with the addition amount from 1 g/kg to 5 g/kg. (b) Histogram of signal intensity of EAd at different viscosity of media, and fitting line with the mass amounts of SCC, Pec, and XG. The concentration of the molecular sensor EAd = 10 μM, λ_{ex} = 320 nm.

influence on the signal emission intensity during the test common pH range. And the pH stability under high viscous glycerol has been tested as well, In addition, the photostability of natural probe EAd among various commercial liquids were tested as well. As shown in Fig. S9, higher stability of fluorescence intensity was maintained under 60 min continuous light irradiation, and no obvious attenuation phenomenon was occurred among these commercial liquids. As presented in Fig. S10, it can be found that the maximum stokes shift of EAd in water varied from 324.9 nm to 434.3 nm, and the stokes shift in glycerol distributed from 329.3 nm to 412.6 nm, respectively. It showed a large stokes shift of 109.4 nm and 83.3 nm, respectively, which may avoid the overlap of absorption the auto-fluorescence and enhance the signal-to-noise ratio (Wang et al., 2024). The results are better than most of the reported studies as listed in Table S1. These data revealed that EAd owns high photostability, excellent chemical stability, and large stokes shift, which suggesting its feasibility of applications in complex liquid system.

3.4. Theoretical calculation

The response mechanism of natural probe EAd to viscosity was further identified, a theoretical calculation based on DFT/B3LYP/6-31G was performed using Gaussian 09 software (Ndaleh et al., 2024). As shown in Fig. S11, the HOMO and LUMO energy levels for ground and excited states of natural probe EAd were measured. The HOMOs were found to be mainly located at phenolic hydroxyl group, while the LUMOs were basically located on at carboxyl group. The energy gaps of natural probe EAd at different rotation states were calculated to be ΔE=6.2159 eV at 0° and ΔE=6.8876 eV, respectively. The HO-MO-LUMO gap in excited state was smaller than that in ground state, which may lead to strong charge transfer and the occurrence of an ICT process. In the high viscous micro-environment, the molecule EAd rotation may be restricted and fluorescence intensity was enhanced due to the transferring between different rotation angels. Moreover, the oscillator strength fem values were calculated to be 0.6538 at 0° and 0.0033 at 90°, the dihedral angels of EAd under excitation state and ground state were quite different, since the conjugate structure of EAd in the excited state was well planarized, and its planarity was destroyed with the rotational motion back to the ground state. These results indicated that intramolecular rotation of EAd was exhibited when the micro-environmental viscosity became lower. Furthermore, the solvatochromism of the natural probe EAd was tested in six kinds of representative solvents, as shown in Fig. 4. Typical red shifting phenomenon in both absorption and emission spectra can be observed, which may be ascribed to the stabilization of ICT process occurring between the phenolic hydroxyl group and carboxyl group (Zheng et al., 2024). These tests confirmed that the intra-molecular rotation of conjugated molecule EAd can form a typical twisted excited state.

3.5. Deterioration process monitoring

To investigate its performance in liquid systems, the natural probe EAd was employed for the detection of viscosity fluctuation in the representative liquid system. Thus, two kinds of commercial liquids named raspberry fruit juice and lemon juice were selected to test its tracking capability during the spoiled process. In detail, these liquids were stored under ambient temperature and lower-maintenance temperature for 7 days, respectively. Corresponding digital images were displayed in Fig. 5a.When the liquids were stored under ambient temperature, at the beginning of two days, both liquids were *trans*-parent and clear appearances, and the color has not dimmed yet. After day 4, the color of raspberry fruit juice has been faded, bleached phenomenon can be observed, especially at day 7. And several kinds of floating objects occurred in the lemon juice and turbid phenomenon became seriously. On the contrary, when these two kinds of liquids were stored under lower-maintenance temperature, a normal appearance and transparent phenomena can be observed even though the storage time was extended to 4 days, as shown in Fig. 5b. After day 4, the apparent color of raspberry fruit juice was still retented, and only muddy phenomena can be found in the lemon juice. During the process of monitoring the deterioration of raspberry fruit and lemon juice, with the increase time of extension storage under ambient temperature and lower temperature, the emission signals have been tracked along the 7 days

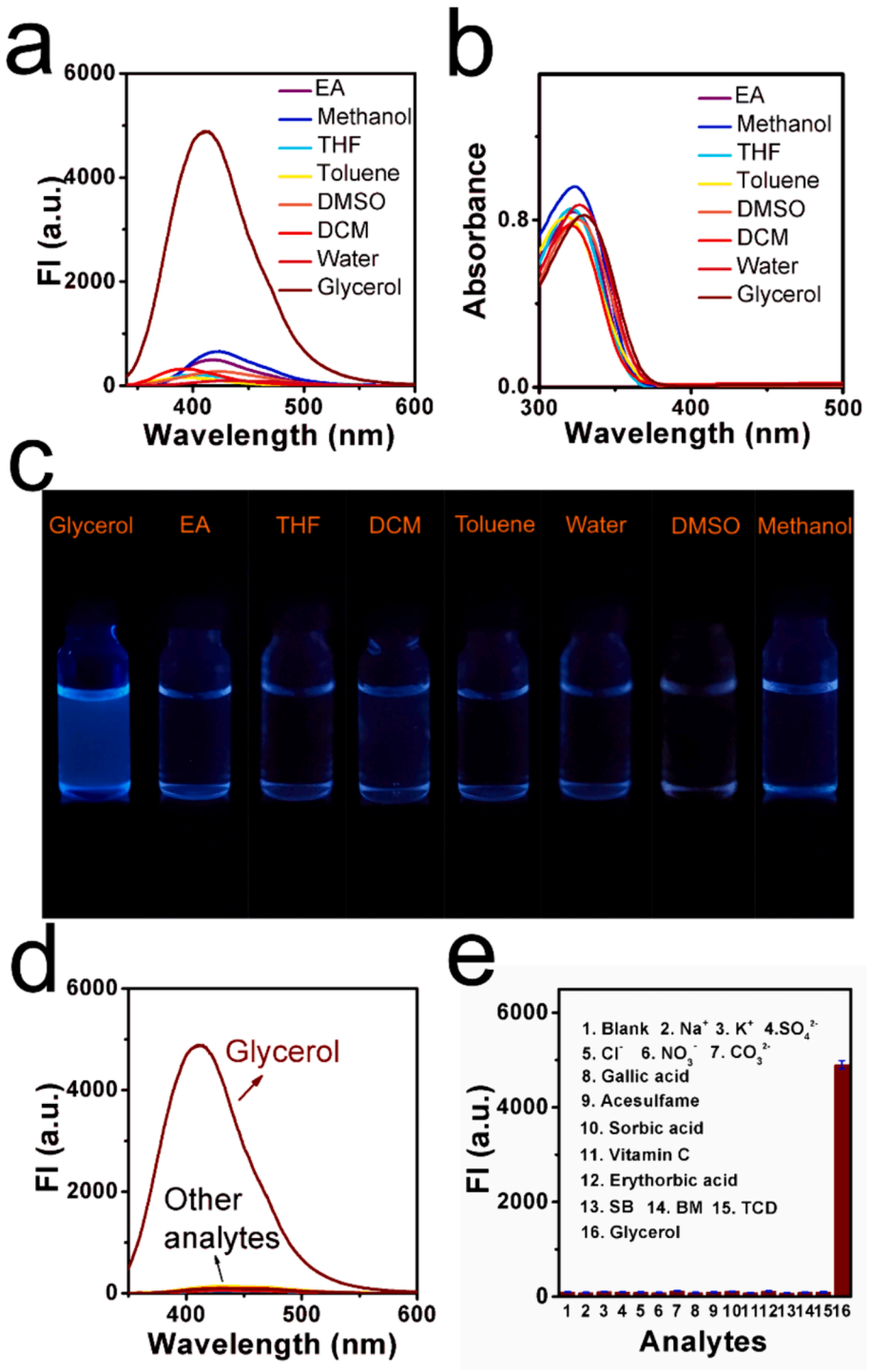


Fig. 3. (a) Fluorescence emissive spectra of the molecular probe EAd in various common solvents. (b) Absorption spectra of the molecular probe EAd in various common solvents with different polarities. (c) Fluorescent images in various solvents. (d) Emission spectra of EAd (10 μM) in various solutions with the potential analytes in liquid food. (e) Selectivity of the probe EAd (10 μM) toward various liquids-related analytes, including blank, Na^+, K^+, Ca^{2+}, SO_4^{2-}, Cl^-, NO_3^-, CO_3^{2-}, gallic acid, acesulfame, sorbic acid, vitamin C, erythorbic acid, sodium benzoate (SB), beet molasses (BM), trisodium citrate dehydrate (TCD), and glycerol. Concentration of the analyte is 50 μM.

with spectrometer. And the whole deterioration process was measured at day 0, day 2, day 4 and day 7. Before the test, all the samples were pretreated with the natural probe EAd with the concentration of 10 μM, and then led to the fluorescence investigation. The results were shown in Fig. 5c and Fig. 5d, it can be found that the FI signal enhanced obviously with the storage time extended, 17.3 % and 20.2 % increment were found after 7 days under ambient temperature in the raspberry fruit and lemon juice, respectively. However, only 9.1 % and 10.6 % enhancements can be found when the raspberry fruit and lemon juice were stored under lower-maintenance temperature. Moreover, both fresh liquids were stored under high temperature (37 °C) as well, as shown in Fig. S12. It can be found that more severe deterioration phenomenon and higher emission enhancements occurred, 21.3 % and 26.9 % enhancements were found in the raspberry fruit and lemon juice, which

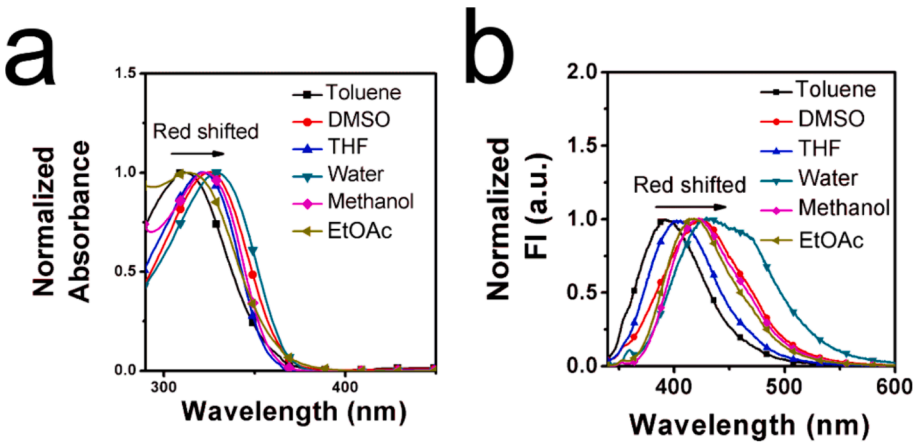


Fig. 4. (a) Normalized absorbance of molecular probe EAd in six presentative solvents. (b) Normalized emission spectra of molecular probe EAd in six presentative solvents.

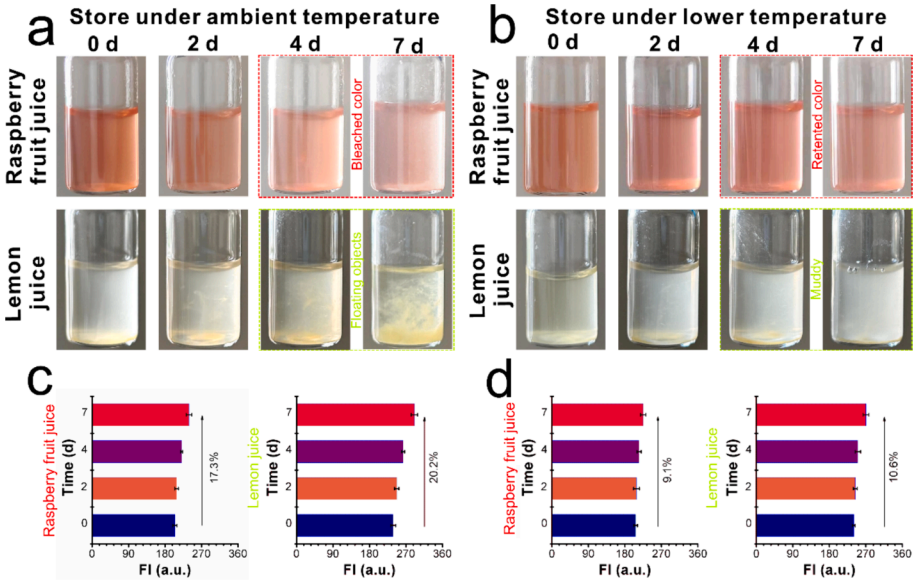


Fig. 5. (a) Digital images of the raspberry fruit juice and lemon juice stored under ambient temperature within 7 days. (b) Digital images of the raspberry fruit juice and lemon juice stored under low-er-maintenance temperature within 7 days. (c) The emission intensity enhancement of raspberry fruit juice and lemon juice during the 7 days under ambient temperature. (d) The emission intensity enhancement of raspberry fruit juice and lemon juice during the 7 days under lower-maintenance temperature. The concentration of EAd = 10 μM, λ_{ex} = 320 nm.

indicated that higher temperature can accelerate the deterioration process. These data confirmed that lower temperature can slow down the deterioration speed, and extend the storage time. More importantly, the abovementioned results indicated that the molecular probe EAd can monitor the micro-environmental viscosity fluctuation, and can be utilized as an out-standing luminescent tool to achieve deterioration extent sensing.

In order to consolidate this conclusion, we studied the change pattern of viscosity values *via* the viscometer as well. As seen from Fig. 6a and Fig. 6b, the viscosities of raspberry fruit and lemon juice increase sharply after day 2 when stored at ambient temperature. 36.4 % and 44.8 % enhancements were tested in the raspberry fruit and lemon juice, respectively. By contrast, the viscosities of raspberry fruit and lemon juice enhanced only 17.7 % and 26.5 % when stored at the lower-maintenance temperature. The results were consistent with above fluorescence intensity results. Notably, a fitting linear relationship was established between the viscosity increment percentage $(\eta_n-\eta_0)/\eta_0 \times 100$ % and the fluorescence intensity increment percentage $(F_n-F_0)/F_0 \times 100$ %, as displayed in Fig. 6c. With the combination of fluorescence spectra and traditional viscosity determination results, it can be concluded that the micro-environmental viscosity became higher during the deterioration process, and normal ones can be distinguished from the spoiled ones through the fluorescent technique.

4. Conclusions

Herein, we have reported one kind of natural erucic acid (EAd) extracted from the rosemary plant, and its viscosity-responsive and emission properties have been investigated, the capability of detecting the viscosity fluctuation during liquid deterioration process has been further explored. Interestingly, this probe EAd displayed higher viscosity efficient (x = 0.56), and an obvious turn-on signal can be found in the high viscosity media. The fluorescence intensity of the probe displayed a strong independence on pH, polarity, and multiple interfering analytes. And the sensitivity, photostability, adaptability, and detect limit have been investigated comprehensively. DFT calculation results explained that the turn-on mode fluorescence at higher viscous media was achieved by the TICT process. Furthermore, this natural probe EAd was

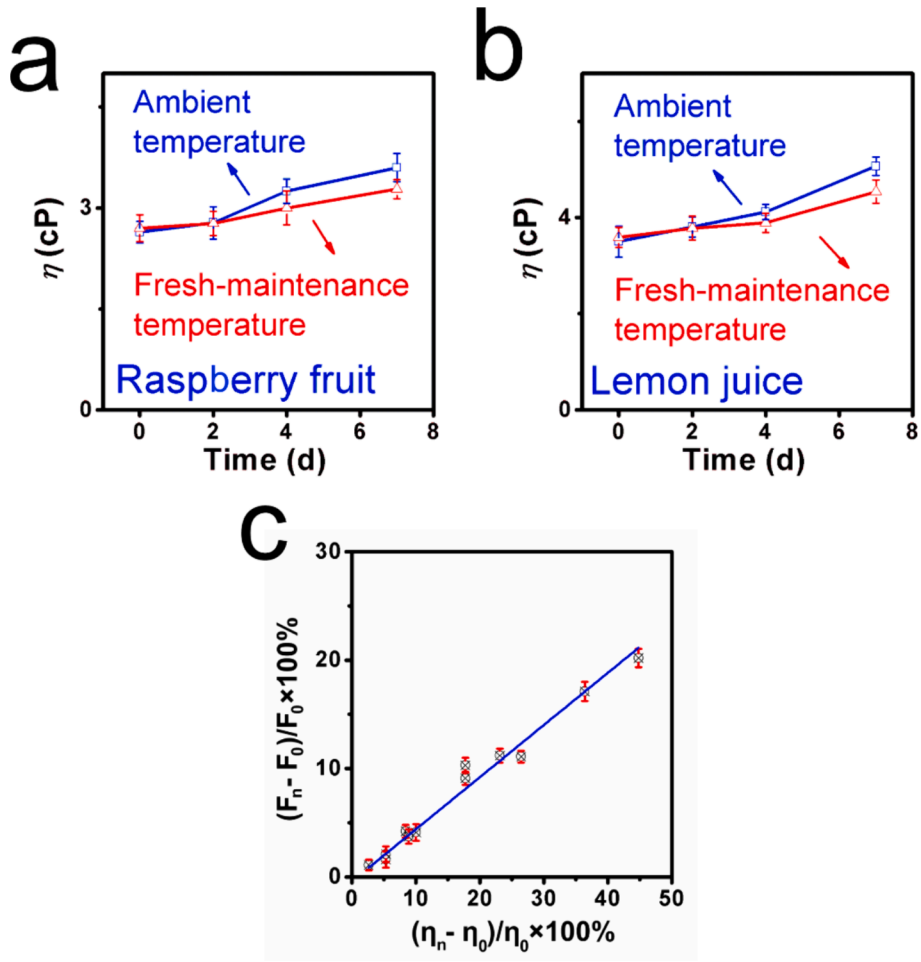


Fig. 6. (a) Viscosity values of raspberry fruit juice when stored under different temperatures. (b) Viscosity values of lemon juice when stored under different temperatures. (c) The fitting line among the fluorescence increment percentage and viscosity enhanced degree.

available for the quantitative determination of thickening effect. And it has been successfully applied to visualized viscosity fluctuations during the liquid deterioration program, positive relationship was found among the metamorphism degree and emission fluorescence signal intensity. The results demonstrated a satisfying fluorescence imaging ability on the liquid safety investigation, in our work, normal fresh liquids can be easily distinguished from the spoiled ones, which validated the ability of the natural probe EAd as the liquid safety visualization tool.

CRediT authorship contribution statement

Lingfeng Xu: Writing – review & editing, Writing – original draft, Resources, Project administration, Methodology, Funding acquisition, Conceptualization. **Lili Zeng:** Software, Investigation, Formal analysis. **Ting Ouyang:** Validation, Investigation, Formal analysis. **Xinmin Deng:** Visualization, Validation, Investigation. **Xinya Liu:** Validation, Investigation, Data curation. **Runlin Han:** Writing – review & editing, Supervision, Funding acquisition.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.arabjc.2024.105954.

References

Chen, Q., Fang, H., Shao, X., Tian, Z., Geng, S., Zhang, Y., Fan, H., Xiang, P., Zhang, J., Tian, X., Zhang, K., He, W., Guo, Z., Diao, J., 2020. A dual-labeling probe to track functional mitochondria–lysosome interactions in live cells. Nat. Commun. 11, 6290. https://doi.org/10.1038/s41467-020-20067-6.

Cox, S., Sandall, A., Smith, L., Rossi, M., Whelan, K., 2021. Food additive emulsifiers: a review of their role in foods, legislation and classifications, presence in food supply, dietary exposure, and safety assessment. Nutr. Rev. 79, 726–741. https://doi.org/10.1093/nutrit/nuaa038.

Du, X., Wang, B., Li, H., Liu, H., Shi, S., Feng, J., Pan, N., Xia, X., 2022. Research progress on quality deterioration mechanism and control technology of frozen muscle foods.

Compr. Rev. Food Sci. Food Saf. 21, 4812–4846. https://doi.org/10.1111/1541-4337.13040.
Gao, R., Wu, Z., Ma, Q., Lu, Z., Ye, F., Zhao, G., 2021. Effects of breaking methods on the viscosity, rheological properties and nutritional value of tomato paste. Foods 10. https://doi.org/10.3390/foods10102395.
Ge, J., Cai, W., Niu, N., Wen, Y., Wu, Q., Wang, L., Wang, D., Tang, B.Z., Zhang, R., 2023. Viscosity-responsive NIR-II fluorescent probe with aggregation-induced emission features for early diagnosis of liver injury. Biomaterials 300, 122190. https://doi.org/10.1016/j.biomaterials.2023.122190.
Hajikhani, M., Lin, M., 2022. A review on designing nanofibers with high porous and rough surface via electrospinning technology for rapid detection of food quality and safety attributes. Trends Food Sci. Technol. 128, 118–128. https://doi.org/10.1016/j.tifs.2022.08.003.
He, S., Joseph, N., Feng, S., Jellicoe, M., Raston, C.L., 2020. Application of microfluidic technology in food processing. Food Funct. 11, 5726–5737. https://doi.org/10.1039/D0FO01278E.
Hou, J.-T., Wang, B., Zou, Y., Fan, P., Chang, X., Cao, X., Wang, S., Yu, F., 2020. Molecular fluorescent probes for imaging and evaluation of hypochlorite fluctuations during diagnosis and therapy of osteoarthritis in cells and in a mouse model. ACS Sens 5, 1949–1958. https://doi.org/10.1021/acssensors.0c00270.
Lee, E., Kim, B., Choi, S., 2020. Hand-held, automatic capillary viscometer for analysis of Newtonian and non-Newtonian fluids. Sens Actuators A Phys 313, 112176. https://doi.org/10.1016/j.sna.2020.112176.
Li, Y.-Y., Hu, J.-L., Wu, J.-R., Wang, Y.-R., Zhang, A.-H., Tan, Y.-W., Shang, Y.-J., Liang, T., Li, M., Meng, Y.-L., Kang, Y.-F., 2024b. Multifunctional fluorescence probe for simultaneous detection of viscosity, polarity, and $ONOO^-$ and its bioimaging *in vitro* and *vivo*. Biosens. Bioelectron. 254, 116233 https://doi.org/10.1016/j.bios.2024.116233.
Li, F., Li, T., Zhao, J., Fan, M., Qian, H., Li, Y., Wang, L., 2024. Unraveling the deterioration mechanism of dough during whole wheat flour processing: a case study of gluten protein containing arabinoxylan with different molecular weights. Food Chem. 432, 137199 https://doi.org/10.1016/j.foodchem.2023.137199.
Liu, W., Wang, T., Wang, L., Wang, Y., Hu, S., Tian, D., 2024. A novel near-infrared fluorescent probe for ultrasensitive and visual detection of mitochondrial viscosity. Spectrochim. Acta A Mol. Biomol. Spectrosc. 304, 123329 https://doi.org/10.1016/j.saa.2023.123329.
Liu, S.-S., Wu, W.-N., Zhao, X.-L., Fan, Y.-C., Wang, Y., Xu, Z.-H., 2023. A dual-emission fluorescence probe for the detection of viscosity and hydrazine in environmental and biological samples. Anal. Chim. Acta 1245, 340867. https://doi.org/10.1016/j.aca.2023.340867.
Liu, J., Zhang, W., Zhou, C., Li, M., Wang, X., Zhang, W., Liu, Z., Wu, L., James, T.D., Li, P., Tang, B., 2022. Precision navigation of hepatic ischemia-reperfusion injury guided by lysosomal viscosity-activatable NIR-II fluorescence. J. Am. Chem. Soc. 144, 13586–13599. https://doi.org/10.1021/jacs.2c03832.
Ma, M., Sun, Q.-J., Li, M., Zhu, K.-X., 2020. Deterioration mechanisms of high-moisture wheat-based food – a review from physicochemical, structural, and molecular perspectives. Food Chem. 318, 126495 https://doi.org/10.1016/j.foodchem.2020.126495.
Ma, X., Zhang, X., Zhang, B., Yang, D., Sun, H., Tang, Y., Shi, L., 2024. Dual-responsive fluorescence probe for measuring HSO_3^- and viscosity and its application in living cells and real foods. Food Chem. 430, 136930 https://doi.org/10.1016/j.foodchem.2023.136930.
Mäkelä, N., Brinck, O., Sontag-Strohm, T., 2020. Viscosity of β-glucan from oat products at the intestinal phase of the gastrointestinal model. Food Hydrocoll. 100, 105422 https://doi.org/10.1016/j.foodhyd.2019.105422.
Miao, R., Li, J., Wang, C., Jiang, X., Gao, Y., Liu, X., Wang, D., Li, X., Liu, X., Fang, Y., 2022. A general method to develop highly environmentally sensitive fluorescent probes and AIEgens. Adv. Sci. 9, 2104609. https://doi.org/10.1002/advs.202104609.
Moradi, M., Razavi, R., Omer, A.K., Farhangfar, A., McClements, D.J., 2022. Interactions between nanoparticle-based food additives and other food ingredients: a review of current knowledge. Trends Food Sci. Technol. 120, 75–87. https://doi.org/10.1016/j.tifs.2022.01.012.
Ndaleh, D., Meador, W.E., Smith, C., Friedman, H.C., McGuire, M., Caram, J.R., Hammer, N.I., Delcamp, J.H., 2024. Near-infrared emissive indolizine squaraine fluorophores as strong molecular viscosity sensors. ChemPhotoChem 8, e202300212. https://doi.org/10.1002/cptc.202300212.
Pan, W., Han, L., Cao, X., Shen, S., Pang, X., Zhu, Y., 2023. Dual-response near-infrared fluorescent probe for detecting cyanide and mitochondrial viscosity and its application in bioimaging. Food Chem. 407, 135163 https://doi.org/10.1016/j.foodchem.2022.135163.
Pirsa, S., Hafezi, K., 2023. Hydrocolloids: Structure, preparation method, and application in food industry. Food Chem. 399, 133967 https://doi.org/10.1016/j.foodchem.2022.133967.
Sabzehmeidani, M.M., Kazemzad, M., 2022. Quantum dots based sensitive nanosensors for detection of antibiotics in natural products: a review. Sci. Total Environ. 810, 151997 https://doi.org/10.1016/j.scitotenv.2021.151997.
Sagdeev, D.I., Gabitov, I.R., Khairutdinov, V.F., Fomina, M.G., Alyaev, V.A., Sal'manov, R.S., Minkin, V.S., Gumerov, V.S., Abdulagatov, I.M., 2020. New Design of the Falling-Body Rheoviscometer for High and Extra-High Viscous Liquid Measurements. Viscosity of Vacuum Oils. J. Chem. Eng. Data 65, 1773–1786. https://doi.org/10.1021/acs.jced.9b01071.
Song, H., Zhang, W., Zhang, Y., Yin, C., Huo, F., 2022. Viscosity activated NIR fluorescent probe for visualizing mitochondrial viscosity dynamic and fatty liver mice. Chem. Eng. J. 445, 136448 https://doi.org/10.1016/j.cej.2022.136448.
Sun, M., Wang, T., Yang, X., Yu, H., Wang, S., Huang, D., 2021. Facile mitochondria localized fluorescent probe for viscosity detection in living cells. Talanta 225, 121996. https://doi.org/10.1016/j.talanta.2020.121996.
Wang, C., Qiao, Q., Chi, W., Chen, J., Liu, W., Tan, D., McKechnie, S., Lyu, D., Jiang, X.-F., Zhou, W., Xu, N., Zhang, Q., Xu, Z., Liu, X., 2020. Quantitative design of bright fluorophores and AIEgens by the accurate prediction of twisted intramolecular charge transfer (TICT). Angew. Chem. Int. Ed. Engl. 59, 10160–10172. https://doi.org/10.1002/anie.201916357.
Wang, H., Sun, Y., Lin, X., Feng, W., Li, Z., Yu, M., 2023. Multi-organelle-targeting pH-dependent NIR fluorescent probe for lysosomal viscosity. Chin. Chem. Lett. 34, 107626 https://doi.org/10.1016/j.cclet.2022.06.049.
Wang, B., Yang, D., Zhong, X., Liu, Y., Huang, Y., 2024. A red-emission fluorescent probe with large stokes shift for detection of viscosity in living cells and tumor-bearing mice. Molecules 29. https://doi.org/10.3390/molecules29091993.
Xiao, H., Li, P., Tang, B., 2021. Small molecular fluorescent probes for imaging of viscosity in living biosystems. Chemistry 27, 6880–6898. https://doi.org/10.1002/chem.202004888.
Xu, S.-L., Guo, F.-F., Xu, Z.-H., Wang, Y., James, T.D., 2023. A hemicyanine-based fluorescent probe for ratiometric detection of ClO- and turn-on detection of viscosity and its imaging application in mitochondria of living cells and zebrafish. Sens. Actuators B 383, 133510. https://doi.org/10.1016/j.snb.2023.133510.
Xu, Q., Ren, M., Liu, K., Wang, X., Wang, J.-Y., Wang, S., Kong, F., 2022. Construction of chitosan-based fluorescent probe for real-time monitoring of viscosity changes in living cells with low cytotoxicity and high photostability. Chem. Eng. J. 430, 132851 https://doi.org/10.1016/j.cej.2021.132851.
Yang, X., Zhang, D., Ye, Y., Zhao, Y., 2022a. Recent advances in multifunctional fluorescent probes for viscosity and analytes. Coord. Chem. Rev. 453, 214336 https://doi.org/10.1016/j.ccr.2021.214336.
Ye, S., Zhang, H., Fei, J., Wolstenholme, C.H., Zhang, X., 2021. A general strategy to control viscosity sensitivity of molecular rotor-based fluorophores. Angew. Chem. Int. Ed. 60, 1339–1346. https://doi.org/10.1002/anie.202011108.
Zhang, Q., Liu, W., Jiang, L., He, Y.-J., Wu, C.-J., Ren, S.-Z., Wang, B.-Z., Liu, L., Zhu, H.-L., Wang, Z.-C., 2024. Real-time monitoring of abnormal mitochondrial viscosity in glioblastoma with a novel mitochondria-targeting fluorescent probe. Analyst 149, 2956–2965. https://doi.org/10.1039/D4AN00226A.
Zhang, X., Zhu, D., Yang, X., Man, C., Jiang, Y., Zhao, Q., Zhang, X., 2024. Nanozyme-enabled microfluidic biosensors: a promising tool for on-site food safety analysis. Trends Food Sci. Technol. 148, 104486 https://doi.org/10.1016/j.tifs.2024.104486.
Zheng, Y., Zhai, S.-M., Xiao, M.-M., Dong, P.-Z., Xu, J.-R., Zhao, B.-X., 2024. A novel ratiometric fluorescence probe based on the FRET-ICT mechanism for detecting fluoride ions and viscosity. Spectrochim. Acta A Mol. Biomol. Spectrosc. 309, 123822 https://doi.org/10.1016/j.saa.2023.123822.
Zhou, C., Ma, J., Sun, D.-W., 2023. Grouping illuminants by aggregation-induced emission (AIE) mechanisms for designing sensing platforms for food quality and safety inspection. Trends Food Sci. Technol. 134, 232–246. https://doi.org/10.1016/j.tifs.2023.03.002.