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A novel method for the determination of fast green in grape wine based on resonance Rayleigh scattering

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ABSTRACT

A novel resonance Rayleigh scattering method was developed for the determination of fast green (FCF) in grape wine. In pH 2.5 Britton Robinson (BR) buffer solution, the scattering signal of acridine orange (AO) was remarkably enhanced after adding trace amount of FCF and forming an ion-association complex, which not only resulted in the change of absorption spectrum, fluorescence spectra, but also led to a significant enhancement of resonance Rayleigh scattering (RRS), frequency doubling scattering (FDS), and second order scattering (SOS). The linear ranges and detection limits for RRS, SOS and FDS were 2 −45 × 10^{-6} mol L^{-1}, 2–24 × 10^{-6} mol L^{-1}, 2–20 × 10^{-6} mol L^{-1}, and 8.0 × 10^{-8} mol L^{-1}, 4.7 × 10^{-7} mol L^{-3}, 1.0 × 10^{-7} mol L^{-3}, respectively. In this work, the optimum conditions, the influencing factors and the effects of coexisting substances on the reaction were investigated. The method can be applied to the determination of FCF in grape wine and the results were satisfactory.

1. Introduction

Edible pigment is a kind of food additives, also known as colorant, they are edible dyes used to improve the look of goods. Commonly used in food processing, beverage, medicine, lipstick and cosmetics of dyeing. The food coloring usually divided into two categories, natural pigment and synthetic pigment. With the development of the society and people living standard rise, more and more people double the synthetic pigment using in food can cause harm to human body health or not. At the same time, large numbers researches pointed out that almost all of the synthetic pigments can not provide nutrients to the human body health, and it has been confirmed that some food additives have carcinogenic, teratogenic and mutagenic effects, particularly if they are excessively consumed [1,2].

Fast green (FCF: Fig. 1), also known as food green 3, it is widely used in candy coating, drinks, ice cream, health care products, skin care products and toothpaste [3]. Food colorings affect the physical and mental health of children, leading them to lose control, cry loudly, develop insomnia and other effects [4]. So, it is necessary to limit the mount of FCF added into food and guarantee the safety of the consumers. For this purpose, some available methods were developed resent years, such as high performance liquid chromatography (HPLC) [5], High resolution nuclear magnetic altogether [6], thin layer chromatography, capillary electrophoresis, LC-MS and GC–MS [7–11]. However, among these methods, disadvantages which include insensitivity and excessive labor cost and time-consuming in sample pretreatment exist in each approach. Compared with other analytical techniques, resonance Rayleigh scattering and fluorescence methods which used dye as probe are generally fast and economical in trace analysis.

Resonance Rayleigh scattering (RRS), a new analytical technique developed in the 1990s [12], it is a phenomenon of elastic light scattering and RRS experiments are usually performed at wavelengths away from absorption bands, but for species that aggregate, enhancements in light scattering of several orders of magnitude can be observed at wavelengths characteristics of these species [13]. Due to the sensitivity, convenience in performance and simplicity of the apparatus required, the RRS technique has been widely used to analysis metal ions [14–16], organics [17], biomacromolecule and pharmaceutics [18–25] in recent years due to its simplicity and sensitivity. At the same time, resonance nonlinear scattering (RNLC), such as frequency doubling scattering (FDS), second order scattering (SOS) attract attention on analysis and get more applications [26–32]. In the past, people always used

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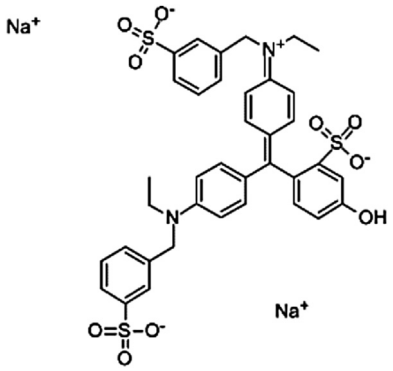

Fig. 1. Molecular structure of FCF.

dyes as RRS probe for the determination of other substance, but using the dyes as RRS probe to detect dye is few reports.

Hence, we reported a method using acridine orange (AO) as RRS and fluorescence probe for the determination of FCF. In this article, we studied the interaction between AO and FCF, it was found that the RRS, SOS, FDS intensity of AO and FCF were very weak, however, when AO interacted with FCF, the RRS, SOS, FDS intensity of the system were remarkably enhance. At the same time, the fluorescence intensity of AO was quenched in the presence of FCF. These methods all could be developed through these experiment and applied to determine FCF.

2. Experimental

2.1. Reagents and apparatus

All reagents used were of analytical grade and used without further purification. Stock solution of AO (1.0×10^{-3} mol L^{-1}) and FCF (1.0×10^{-3} mol L^{-1}) were prepared and maintained at 4 °C. Working solutions were freshly prepared by diluting the corresponding stock solution. Britton–Robinson (BR) buffer solution with different pH were prepared by mixing the mixed acid (H_3PO_4, HAc, H_3BO_3) and 0.2 mol L^{-1} NaOH in different proportions.

A Hitachi F-2500 spectrofluorophotometer (Hitachi Company, Tokyo, Japan) was used to record the fluorescence spectra and RRS, SOS, FDS spectra. A UV-2450 spectrophotometer (Tianmei Corporation, Shanghai, China) was applied to record the absorption spectra. A PHS-3C pH meter (Leici, Shanghai, China) was used to adjust the pH values of the aqueous solutions.

2.2. General procedure

To a 5 mL calibrated flask were added 0.5 mL BR buffer solution (pH 2.5), 0.6 mL 2.0×10^{-4} mol L^{-1} AO solution and suitable amounts of FCF, then diluted with deionized water to the mark and mixed thoroughly with gentle shake. The solution mixture was then incubated at room temperature for 10 min. The RRS, SOS, FDS, fluorescence and absorption spectra were measured.

3. Results and discussion

3.1. RRS spectra

From Fig. 2A, we knew that the RRS signals of FCF and AO were both weaker. AO had three resonance Rayleigh scattering signal near 245 nm, 287 nm and 333 nm, respectively. With the addition of FCF, stronger RRS peaks occurred at both 287 nm and 333 nm. The two RRS peaks (287 nm and 333 nm) in the RRS spectra was on the right of corresponding absorption peak (as shown in the inset of Fig. 5), this was a characteristic "absorption-scattering" phenomenon of RRS spectra. Compared with RRS signal of AO, RRS signal at 333 nm was stronger wavelength, it may not only avoid the adverse reaction from the higher radiant energy of short wavelength, but also reduced the background interference and accordingly increase the RRS strength. Therefore, this experiment adopted $\lambda em = \lambda ex = 333$ nm as the study wavelength. The RRS intensity of AO in the presence of various concentration of FCF was shown in Fig. 2. It can be seen that under the optional experimental conditions, the RRS intensity of AO (curve a) and FCF (curve b) were rather weak. However, when AO reacted with FCF, the RRS intensity enhanced remarkably and a new RRS spectra appears, and the maximum scattering peak is locked at 333 nm (Fig 2A). At the same time, the enhancement of RRS intensity was in proportional to the concentration of FCF in the range of $2–45 \times 10^{-6}$ mol L^{-1} (Fig 2B).

Fig. 3A and B shown the SOS and FDS spectra of AO-FCF system. The maximum wavelengths of SOS, FDS were located at 650 nm and 331 nm, respectively. Under experiment conditions, the SOS and FDS intensity of AO (curve a) and FCF (curve b) were very weak, after AO interacted with FCF, the intensity of SOS and FDS can be enhanced, and the enhancement of intensity were in proportion to the concentration of FCF in the range of $2–24 \times 10^{-6}$ mol L^{-1}, $2–20 \times 10^{-6}$ mol L^{-1}, respectively.

3.2. Fluorescence spectra

Fig. 4 shows the fluorescence quenching of AO upon interaction with different concentration of FCF under the optimal conditions. As the concentration of FCF increased, the quenching of AO was enhanced. Calibration curve in the inset of Fig. 4 were the plots of the optical analysis of different concentration of FCF by the quenching of the emission maximum of AO. The results exhibited a good linear relationship in the range of $2–16 \times 10^{-6}$ mol L^{-1}.

3.3. Absorption spectra

Under the optimum conditions, the absorption spectra of AO, FCF and their complexes were researched, shown in the inset of Fig. 5 The results demonstrated that the absorption of AO is located at 468 nm, and the maximum absorption wavelength of FCF was at 622 nm. When AO reacted with FCF, the maximum absorption of AO, FCF at 468 nm and 622 nm was decreased, respectively. And the absorption intensity was directly proportional to the concentration of FCF in certain range (Fig. 5).

3.4. Optimum reaction conditions

3.4.1. Effect of acidity

The influences of solution acidity on the RRS intensity of the reaction system were tested. As shown in Fig. 6A, it was observed that by keeping the AO and FCF concentrations constant, while changing the pH of BR buffer solution. We found that the enhanced intensity (ΔI_{RRS}) reached the highest at pH 2.5, when the acidity was higher or lower than the optimum point, ΔI_{RRS} decreased gradually. That is because in this work, FCF interacted with AO through electrostatic interaction, if the pH of the system is too higher, it is not good for AO protonize and turn into positive charged. And if the pH is too lower, it may be bad for the existence of negative charged FCF. Hence, when the acidity was higher or lower than the optimum range, ΔI_{RRS} decreases gradually, therefore, subsequent studies were performed at pH 2.5.

3.4.2. Effect of AO concentration

The effect of AO concentration on the RRS intensity was