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# A novel method for detecting allura red based on triple-wavelength overlapping resonance Rayleigh scattering

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**Abstract** A method was presented for sensitive and selective determination of trace allura red (AR) with ethyl violet (EV) in drink samples based on triple-wavelength overlapping resonance Rayleigh scattering (TWO-RRS). At pH 10.0 Britton-Robinson (BR) buffer medium, AR combined with EV to form ion-association complex, which resulted in the RRS intensity enhanced significantly with the new RRS peaks appearing at 341, 508 and 666 nm. The scattering intensities of the three peaks were proportional to the concentration of AR in the ranges of 0.057-5.0 μmol $L^{-1}$ (0.028-2.48 μg $mL^{-1}$). The detection limits for the three single peaks were 0.048 μmol $L^{-1}$ (0.024 μg $mL^{-1}$), 0.050 μmol $L^{-1}$ (0.025 μg $mL^{-1}$), and 0.057 μmol $L^{-1}$ (0.028 μg $mL^{-1}$), while that of the TWO-RRS method was 0.017 μmol $L^{-1}$ (0.008 μg $mL^{-1}$), indicating that the TWO-RRS method could detect trace AR with high sensitivity. In addition, the optimum reaction conditions and the effects of foreign substances were studied, the composition of the ion-association complex and the reasons for the enhancement of RRS were also investigated. The proposed method was successfully applied in real sample analysis with satisfactory results.

**Graphical abstract** A simple, sensitive and selective assay was established to detecting trace allura red in drinks with ethyl violet based on triple-wavelength overlapping resonance Rayleigh scattering.



AR: ● EV: ◖ AR-EV complex:
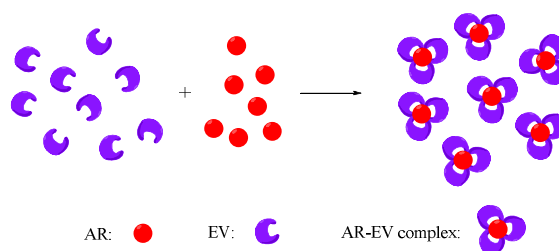
**Key words:** Resonance Reyleigh Scattering; Triple-wavelength Overlapping; Allura Red; Ethyl Violet

## 1. Introduction

Recently, an increasing consideration is focused on food safety due to a series of health risks induced by food. Many diseases spread through food, and food safety involves ways in dealing with, preparing, and storing food to avoid food poisoning, which include safety between the industry and the market that refers to food labeling, food hygiene, food additives and pesticide residues, along with a series of policies for food and its import and export inspection as well as certification systems, and then between the market and the consumer that relating to food should be safe in the market and for the consumer.[1] In considering food safety, food additive is one of the most serious problems since they are prevalent to make food more attractive, but most food additives are synthetic chemicals, toxic to varying degrees. It is confirmed that some food additives have carcinogenic, teratogenic and mutagenic effects, particularly if they are excessively consumed.[2]

Allura red, disodium 6-hydroxy-5-(2 methoxy-5-methyl-4-sulphophenylazo)-2-naphthalenes -ulphonate (Fig. 1), is a food colourant, bright crimson, widely used in candy coating, ice cream, candy, cakes, drinks and other coloring.[3] Food colorings not only affect the physical and mental health of children, leading them lose control, crying loudly, insomnia and other states, but also have some impact on adults with allergies, such as making them appear irritability unstable emotion.[4] Due to this, some regulations have been established to limit the amount of AR added into food to guarantee the consumers' safety. In China, the maximum allowable amount of AR used in drinks is 0.1 g kg$^{-1}$ according to GB2760-2011.[5] A mountain of work were done to develop various techniques including spectrophotometry,[3,6] electrochemical techniques,[7,8] HPLC,[9-11] capillary electrophoresis,[12,13] to determine AR. Spectrophotometry method[3,6] for detecting AR has high sensitivity, and the detection limit are 0.016 μmol L$^{-1}$ and 0.005 μmol L$^{-1}$. Yet it is time-consuming, since it needs separation and preconcentration before determination. Electrochemical technique is used since AR belongs to azo dye, and contain –N=N– group, which is electrochemically active and can be reduced at the mercury electrode. However, mercury electrode is toxic, bringing about environmental pollution and severe adverse effects to human being. Even though some of these methods use a modified electrode to replace the mercury electrode prior to the analysis, but modifications make the analysis cumbersome. Chromatographic methodology can be employed in simultaneous multi-component detection and possesses high separation efficiency, especially liquid chromatography-mass spectrometry (LC-MS) also have been applied to the fast detection of food additives in foodstuffs. Yet equipments are expensive and the analysis cost is high. Capillary electrophoresis is a valuable method owing to its high efficiency, low waste production and fast separation. Nevertheless, the repeatability is not very satisfactory. Developing a novel and advancing technique for detecting AR is therefore urgently needed.

Resonance Rayleigh scattering (RRS) as a well-developed analytical technique has attracted overwhelming attention in the last few years owing to its high sensitivity and simplicity,[14] which has been widely employed in detecting some inorganic ions,[15-17] organic compounds,[18] surfactants,[19,20] pharmaceuticals,[21-24] and macromolecules such as nucleic acids, saccharides and proteins.[25-27] Although the RRS technique has the advantage of practical ease, high sensitivity, and simplicity to investigate aggregate systems, some defects still exist due to the output signals are generally single-wavelength responses, which suffer from variable factors such as instrument

123    response, probes concentration, environment around the probes, and other variable factors.
124    Recently, Hao et al. reported a triple-wavelength overlapping resonance Rayleigh scattering
125    (TWO-RRS) method for the detection of nano-gram dextran sulfate,[28] Zhu et al.[29,30] and Cui et
126    al.[23], respectively, developed a dual-wavelength overlapping resonance Rayleigh scattering
127    (DWO-RRS) method for drug analysis. These multi-response RRS techniques proved much higher
128    sensitivity than the single-wavelength method. Multi-response RRS as a novel and improved
129    technique is a revolutionary milestone in the development of RRS although its generating
130    mechanisms remain abstruse and a few efforts is currently focused on it. Moreover, it could be a
131    great potential tool in promoting long-term development of RRS technique.

132    Even though RRS technique had been employed in detecting some food colorants such as
133    amaranth[31] and erythrosine[32]. Triple-wavelength overlapping resonance Rayleigh scattering
134    method for food colorants assay has not been reported previously, here in this paper a
135    triple-wavelength overlapping resonance Rayleigh scattering method was first established for AR
136    assay with ethyl violet (EV, Fig. 1).



137    AR    EV
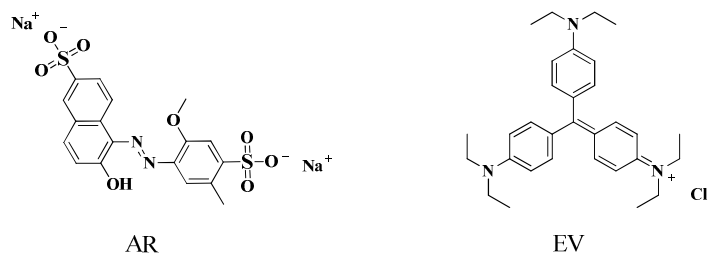
138    **Fig. 1**. The chemical structure of AR and EV

## 2. Experimental

### 2.1 Instrumentation

141    A Hitachi F-2500 spectrofluorophotometer (Tokyo, Japan) was employed for recording
142    scattering spectra and measuring the scattering intensities. A UV-2450 spectrophotometer
143    (Shimadzu, Japan) was used for acquiring absorption spectra and measure absorbance. A pHS-3D
144    pH meter (Shanghai Scientific Instruments Company, China) was used for adjusting the pH
145    values.

### 2.2 Reagents

147    A stock solution of AR ($1.0 \times 10^{-4}$ mol L$^{-1}$) and EV ($2.0 \times 10^{-4}$ mol L$^{-1}$) was prepared and
148    kept at 4 $^{\circ}$C. Working solutions were freshly prepared by diluting the corresponding stock solution.
149    Britton-Robinson (BR) buffer solutions with different pH were prepared by mixing the mixed acid
150    (composed of 2.71 mL 85% $H_3PO_4$, 2.36 mL HAc and 2.47 g $H_3BO_3$) with 0.2 mol L$^{-1}$ NaOH in
151    different proportions. The buffer solutions were used to control the acidity. All reagents were of
152    analytical reagent grade and were used without further purification. Doubly distilled water was
153    used throughout.

### 2.3 General procedure

155    Into a 10 mL calibrated flask were added 1.0 mL of pH 10.0 BR buffer solution, 0.8 mL of
156    $2.0 \times 10^{-4}$ mol L$^{-1}$ EV solution and suitable amounts of AR solution in turn. Then the mixture was
157    diluted to the mark with doubly distilled water and thoroughly mixed at room temperature (25 ± 5
158    $^{\circ}$C). After 20 min, record the RRS spectra of the systems with synchronous scanning at $\lambda_{ex} = \lambda_{em}$ ($\Delta$

159    $\lambda$= 0 nm) and measure the intensity $I$ of the complex and $I_0$ of the reagent blank respectively, $\Delta I =$
160    $I - I_0.$

## 3. Results and discussion

### 3.1 RRS spectra

163    The RRS spectra of AR, EV and their complex at pH 10.0 from 220 to 800 nm were shown
164    in Fig. 2. It can be seen that under the optimum conditions, the RRS intensities of AR or EV were
165    rather weak and AR with only one peak (370 nm) and EV with three peaks (290, 349 and 654nm)
166    in the whole scanning wavelength region. However, when AR reacted with EV to form an
167    ion-association, RRS spectra enhanced remarkably and a new spectrum appeared with three peaks
168    located at 341, 508 and 666 nm, which indicated that the interaction between AR and EV had
169    really occurred. All these peaks had linear relation with the concentration of AR (Fig. 1S), which
170    suggested that the system can be applied to detect AR.
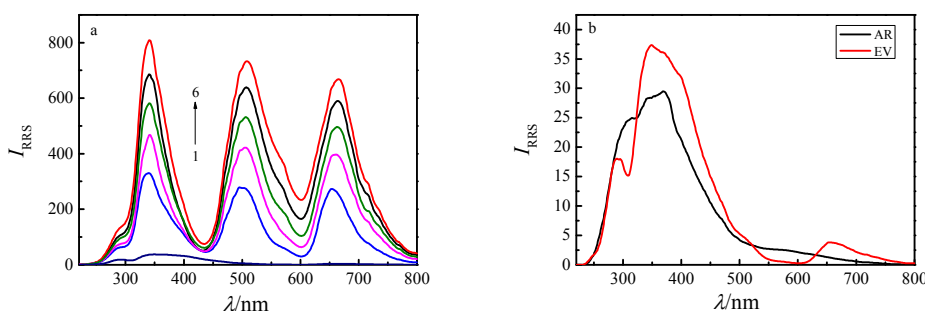
171
172    **Fig. 2**. RRS spectra. (a) RRS spectra of AR-EV system. (1-6) $C_{EV}$ = 1.6 × 10$^{-5}$ mol L$^{-1}$, $C_{AR}$ = 0, 1.0 × 10$^{-6}$, 2.0 ×
173    10$^{-6}$, 3.0 × 10$^{-6}$, 4.0 × 10$^{-6}$, 5.0 × 10$^{-6}$ mol L$^{-1}$, pH 10.0. (b) RRS spectra of AR and EV. AR ($C_{AR}$ = 5.0 × 10$^{-6}$ mol
174    L$^{-1}$), EV ($C_{EV}$ = 1.6 × 10$^{-5}$ mol L$^{-1}$), pH 10.0.

### 3.2 Optimum experimental conditions

176    The experiment indicated that fluctuating trend of the RRS intensity of the three peaks were
177    uniform in all kinds of influencing factors. In this work, 341 nm was selected as the determination
178    wavelength in further experiments.

### 3.2.1 Effect of acidity

180    Fig. 3 indicated the influences of the solution acidity on the RRS intensities of the system
181    with BR buffer. It was observed by keeping the AR and EV concentrations constant while
182    changing the pH of BR buffer solution. We found that the enhanced intensities ($\Delta I$) reached the
183    highest and keep constant at pH range of 9.6-10.4. When the acidity was higher or lower than the
184    optimum range, $\Delta I$ decreased gradually. Therefore, subsequent studies were performed at pH 10.0.
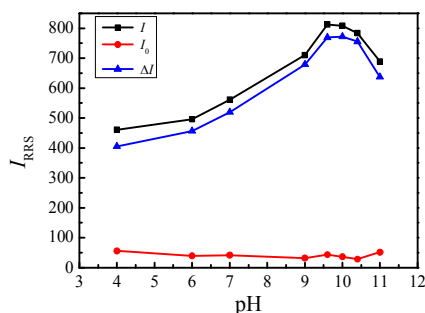
185

186        **Fig. 3.** Effects of acidity. $C_{AR} = 5.0 \times 10^{-6}$ mol L$^{-1}$, $C_{EV} = 1.6 \times 10^{-5}$ mol L$^{-1}$.

187

188    **3.2.2 Effect of EV concentration**

189        The effect of EV concentration on the RRS intensities was investigated by keeping the

190    concentration of AR ($5.0 \times 10^{-6}$ mol L$^{-1}$) constant while varying the concentration of EV and the

191    experiment results showed that the optimum EV concentration was $1.6 \times 10^{-5}$ mol L$^{-1}$. If the

192    concentration was lower than this value, the RRS intensity decreased owing to the incomplete

193    reaction between AR and EV. However, if the concentration was higher than this value, the RRS

194    intensity also reduced. So $1.6 \times 10^{-5}$ mol L$^{-1}$ was chose for the assay.

195    **3.2.3 Reaction speed and the stability of the system**

196        The reaction speed and the stability of the system were also studied. According to the

197    experimental results, the reaction was completed within 20 min and the RRS intensity remained

198    stable for 50 min, then, with the passing time, $\Delta I$ decreased gradually (Table 1S). The reaction was

199    rapid at room temperature and the influence of temperature between 10 and 50 $^\circ$C on RRS

200    intensity was little (Table 2S). Therefore, the experiment was performed at room temperature.

201    **3.2.4 Effect of ionic strength**

202        The effect of ionic strength on AR-EV system was tested by changing the concentration of

203    NaCl. As it can be seen in Fig. 4, $\Delta I$ had little variation along with increasing the concentration of

204    NaCl from 0.00 to 0.1 mol L$^{-1}$, that is, relative error was less than $\pm$ 5%. Therefore, the scattering

205    signals could relatively keep stable within the ionic strength range of 0.00-0.1 mol L$^{-1}$.

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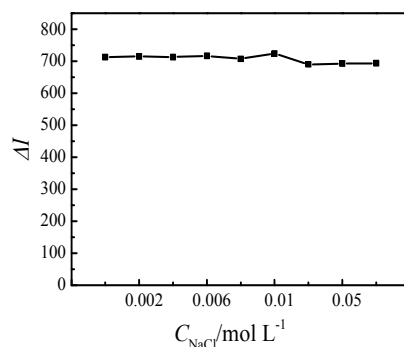
208        **Fig. 4**. Effect of ionic strength. $C_{AR} = 5.0 \times 10^{-6}$ mol L$^{-1}$; $C_{EV} = 1.6 \times 10^{-5}$ mol L$^{-1}$; pH 10.0.

209

210    **3.3 Selectivity of the method**

211        Under the optimum experimental conditions, the influences of foreign coexisting substances

212    on the determination of AR were investigated. As shown in Table 1, when the concentration of AR

213    was $3.0 \times 10^{-6}$ mol L$^{-1}$, a certain amount of common metal ions, some non-metallic ions, sugars,

214    amino acids and some vitamins in drinks can be allowed with relatively high concentration. Some

215    metal ions interfered with the determination, this attributed to the fact that the reaction medium

216    was alkaline. All the interference, however, can be dispelled with the addition of EDTA. Thus this

217    assay presented excellent selectivity and can be applied to the determination of AR in drinks.

218

219

220

221                                    **Table 1** Effects of foreign substances

| Foreign substance | Concentration ($\mu g\ mL^{-1}$) | Relative error (%) | Foreign substance | Concentration ($\mu g\ mL^{-1}$) | Relative error (%) |
|---|---|---|---|---|---|
| KI | 664 | 1.4 | $KAl(SO_4)_2$ | 20 | -3.7 |
| NaCl | 5850 | -3.4 | $NH_4Fe(SO_4)_2$ | 2.5, 50* | 4.9, -4.3* |
| $NH_4Cl$ | 161 | 1.8 | Glucose | 793 | 4.4 |
| $MgSO_4$ | 120 | 3.2 | Sucrose | 1369 | 4. 8 |
| $CaCl_2$ | 16 | 4.8 | Malt sugar | 360 | 1.4 |
| $CuSO_4$ | 2.4, 320* | 4.2, 2.1* | Vitamin B1 | 60 | 2.1 |
| $NiSO_4$ | 3.1, 124* | 4.8, 3.1* | Ascorbic acid | 176 | 2.4 |
| $MnSO_4$ | 3.0, 121* | 2.7, 3.2* | Citric acid | 192 | 1.5 |
| $CdCl_2$ | 3.7, 183* | 3.0, 3.4* | Sodium benzoate | 144 | 2.5 |
| $ZnCl_2$ | 2.5, 272* | 2.6, 1.0* | L-Tryptophan | 163 | 3.2 |
| $(NH_4)_2Fe(SO_4)_2$ | 2.0, 30* | 4.9, 1.3* | L-Arginine | 697 | 1. 9 |
| $Pb(Ac)_2$ | 3.3, 65* | 4.3, 2.9* | L-Aspartic acid | 532 | -2.02 |

222                                    * Added $0.05 mol\ L^{-1}$ EDTA 1.0 mL

223

224     **3.4 Calibration curve**

225         According to the procedure, the scattering intensities of the AR-EV complex were measured.
226     Calibration graphs of $\Delta I$ against the concentrations of AR were depicted in Fig. 5. Table 2
227     displayed the results of single-wavelength (SW-RRS) and three-wavelength overlapping
228     resonance Rayleigh scattering (TWO-RRS) methods. TWO-RRS method showed a higher
229     sensitivity. Additionally, the results of the comparison of several published methods with the
230     proposed method were listed in Table 3. As depicted in Table 2 and Table 3, in response to these
231     previously methods for AR detection, TWO-RRS should be relatively simple, fast and high
232     sensitivity. Our approach offers interesting possibilities to become a new and rapid method for
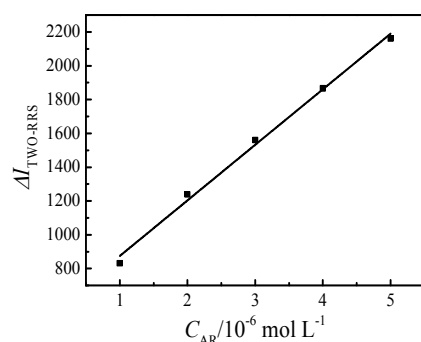233     detecting AR.

234

235                  **Table 2** Analytical parameters of SW-RRS and TWO-RRS methods

| Method | $\lambda$ (nm) | Linear equation (c, $\mu mol\ L^{-1}$) | Linear range (c, $\mu mol\ L^{-1}$) | R | Detection Limit ($3\sigma$, $\mu mol\ L^{-1}$) |
|---|---|---|---|---|---|
| RRS | 341 | $\Delta I=184.3+117.4C$ | 0.160-5.0 | 0.9973 | 0.048 |
| RRS | 508 | $\Delta I=175.9+113.1C$ | 0.166-5.0 | 0.9907 | 0.050 |
| RRS | 666 | $\Delta I=185.7+98.3C$ | 0.191-5.0 | 0.9907 | 0.057 |
| TWO-RRS | 341+508+666 | $\Delta I=545.9+328.8C$ | 0.057-5.0 | 0.9942 | 0.017 |

236

237 **Fig .5**. Calibration graph of AR-EV system. $C_{EV} = 1.6 \times 10^{-5}$ mol L$^{-1}$, $C_{AR} = 1.0 \times 10^{-6}$, $2.0 \times 10^{-6}$, $3.0 \times 10^{-6}$, 4.0
238 $\times 10^{-6}$, $5.0 \times 10^{-6}$ mol L$^{-1}$, pH 10.0.

239

240 **Table 3** Analytical features of some typical methods employed for AR determination

| Method | Linearity ( μmol L$^{-1}$) | Detection limit ( μmol L$^{-1}$) | Remarks |
|---|---|---|---|
| Spectrophotometry[6] | 0.017-12 | 0.005 | High sensitivity and accuracy, need separation and preconcentration before determination. |
| Electrochemical techniques[8] | 0.167-1.2 | 0.050 | Simple and selectivity, but mercury electrode is toxic, bringing about environmental pollution, using a modified electrode to replace the mercury electrode make the analysis cumbersome. |
| HPLC[9] | 0.060-200 | 0.020 | Simultaneous multi-component detection and possesses high separation efficiency, yet equipments are expensive and the analysis cost is high. |
| capillary electrophoresis[13] | 0.140-10.1 | 0.042 | High efficiency, low waste production and fast separation. However, the repeatability is not very satisfactory. |
| TWO-RRS | 0.057-5.0 | 0.017 | Simple, rapid and high sensitivity and selectivity. No pollution for the environment. |

241

242 **3.5 Analytical application of the method**

243 The TWO-RRS method was used to detecting AR in real samples. The drink sample was
244 purchased from a local supermarket, and used directly without any treatment. 1.0 mL of pH 10.0
245 BR buffer solution, 0.8 mL of $2.0 \times 10^{-4}$ mol L$^{-1}$ EV solution and 1.0 mL of the drink sample
246 (without adding EDTA) was added into a 10 mL calibrated flask in turn and diluted to the mark
247 with doubly distilled water and thoroughly mixed. The mixture was detected as the procedure
248 mentioned in section 2.3. Then the recovery was determined by standard addition method.
249 Furthermore, ultraviolet spectrophotometry[3] was conducted to validate the accuracy of TWO-RRS
250 method. The corresponding results were listed in Table 4. It occurred to us that the proposed assay
251 exhibited well accurate (recovery was between 96.6% and 107.1%) and repeatability (RSD was
252 between 2.4% and 4.4%) and the results were in accordance with the literature method. Thus, the
253 proposed method was successfully applied to the determination of AR in soft drink in support of
254 the sensitivity and selectivity of the method to real sample analysis.

255

256                                              **Table 4** Determination of AR in soft drink

| Samples | Found ($\mu$g mL$^{-1}$) | | Added ($\mu$g mL$^{-1}$) | Total found ($\mu$g mL$^{-1}$) | | Recovery (%) | | RSD (%, n=5) | |
|---|---|---|---|---|---|---|---|---|---|
| | TWO-RRS | UV[a] | | TWO-RRS | UV | TWO-RRS | UV | TWO-RRS | UV |
| 1 | 9.4 | 9.2 | 14.9 | 23.8 | 23.9 | 96.6 | 98.7 | 2.4 | 3.2 |
| 2 | 9.4 | 9.2 | 9.9 | 20.0 | 20.1 | 107.1 | 110.1 | 2.6 | 2.1 |
| 3 | 9.4 | 9.2 | 5.0 | 14.5 | 14.1 | 102.0 | 98.0 | 4.4 | 2.8 |

257        [a] Ultraviolet spectrophotometry[3]

258     **3.6 Reaction mechanism and the reasons for RRS enhancement**

259     **3.6.1 Interaction of allura red with ethyl violet**

260          Molar ratio method and Job's method were employed to investigate the composition ratio of

261     AR with EV. The results obtained from the fig. 6 indicated that the ratio of AR and EV is 1 : 3. In

262     aqueous solution, AR can exist as $AR^{2-}$ and $AR^{3-}$, and the distribution ratio depending on the

263     solution acidity. According to the pKa value of AR (pKa=11.4[33]), hydroxyl of AR dissociated

264     rarely (3.8%) and mainly existed as a bivalent anion while EV existed as a univalent cation in pH

265     10.0. Hence 1 mol AR reacted with 2 mol EV to form an ion-association via the electrostatic

266     attraction and hydrophobic force in theory. The formation of the ion-association, however,

267     changed the electron cloud density of AR, which may make hydroxyl of AR dissociate more easily.

268     And nitrogen atom with one positive charge in EV had a strong electron-withdrawing effect,

269     which may cause the combination to oxygen atom of the hydroxyl resulting in dissociation of

270     hydrogen from hydroxyl. Thus, this maybe the reason why the composition ratio of AR with EV in

271     ion-association is 1 : 3 rather than 1 : 2. The interaction between AR and EV was shown in

272     Scheme 1, which not only aroused the remarkable enhancement of RRS, but also caused the

273     changes of absorption spectra. It can be seen from Fig. 7 that AR had a very faint absorption while

274     EV had a very strong absorption peak at 593 nm, after AR was mixed with EV, the absorption

275     peak at 593 nm decreased sharply, which suggested that a new complex had formed.
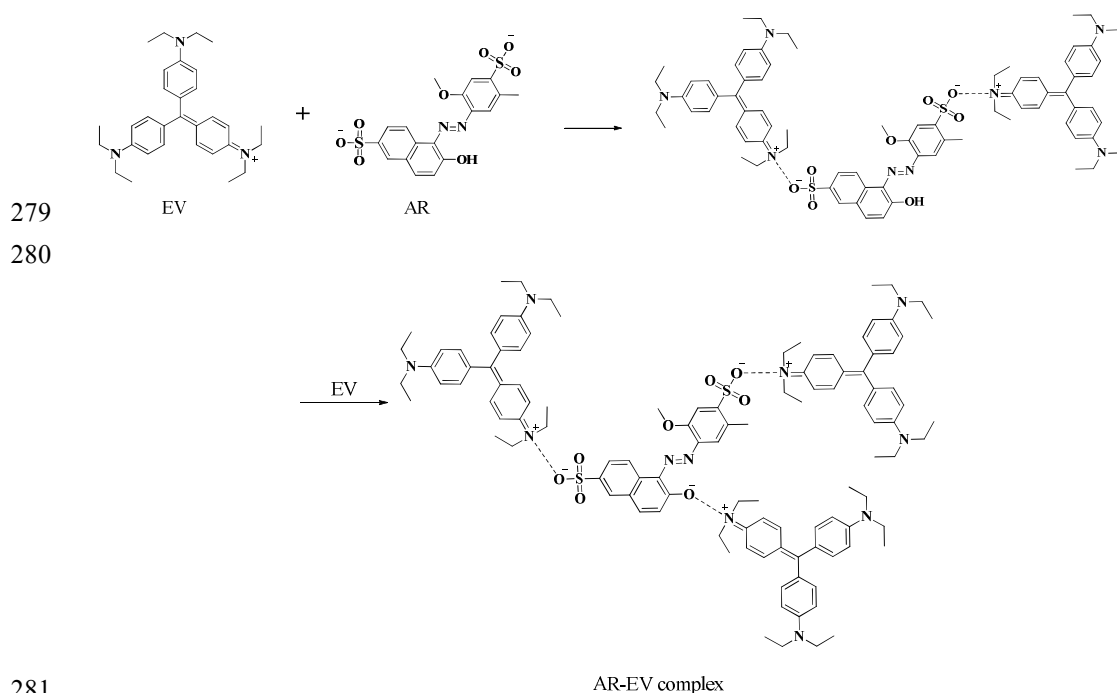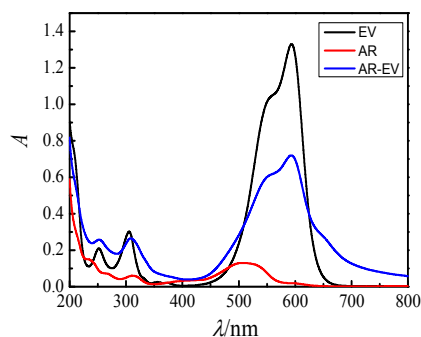


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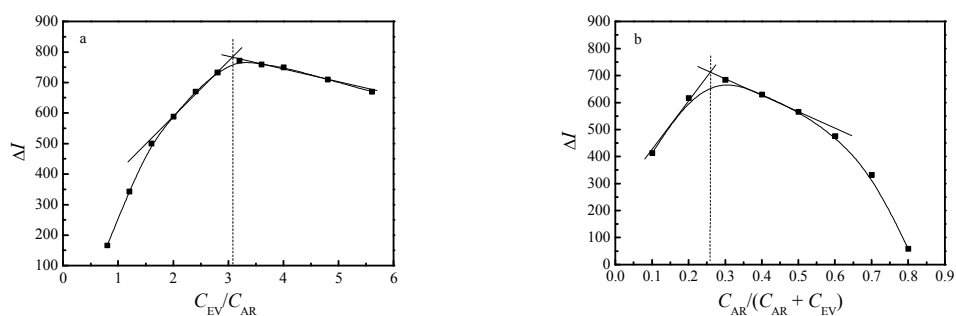
277     **Fig. 6**. The composition ratio of AR with EV. (a). Molar ratio method $C_{AR} = 5.0 \times 10^{-6}$ mol L$^{-1}$, pH = 10.0. (b).

278                          Job's method. $C_{AR} + C_{EV} = 2.0 \times 10^{-5}$ mol L$^{-1}$, pH = 10.0.

279     EV                          AR

280



281                                          AR-EV complex

282                        **Scheme 1**. The associated reaction between AR and EV.



283

284        **Fig. 7**. Absorption spectra. $C_{AR} = 5.0 \times 10^{-6}$ mol L$^{-1}$; $C_{EV} = 1.6 \times 10^{-5}$ mol L$^{-1}$; pH 10.0.

285     **3.6.2 The reasons for RRS enhancement**

286     (1) Resonace enhanced Rayleigh scattering effect

287         When the wavelength of Rayleigh scattering was located at or was close to its absorption

288     band, the same frequency of the electromagnetic wave absorbed by the electron and that of

289     scattering gave birth to the resonance between the Rayleigh scattering and the light absorption.[24]

290     This process tremendously enhanced the scattering intensity and the resonance Rayleigh scattering

291     generated. Thus, it is certain that the RRS spectral characteristics were related to the absorption

292     spectra. Fig. 8 compared the RRS spectrum of the AR-EV complex with its absorption spectrum.

293     It is clear that the RRS peaks (341, 508 and 666 nm) were close to the absorption peak (308 and

294     593 nm).

295
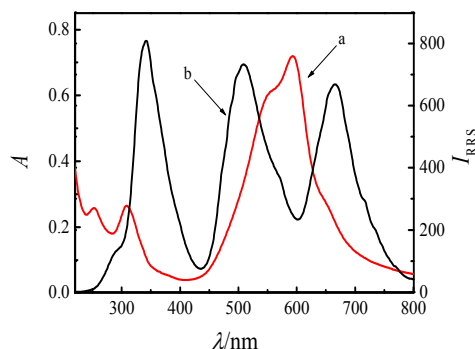
**Fig. 8**. The comparison of absorption spectrum (a) and RRS spectrum (b). $C_{AR} = 5.0 \times 10^{-6}$ mol L$^{-1}$; $C_{EV} = 1.6 \times 10^{-5}$ mol L$^{-1}$; pH 10.0.

298   (2) Hydrophobic effect

299        AR or EV alone can easily dissolved and stably exist as ion in basic solution. After mixed,
300   their charges were neutralized and they lost hydrophilicity and appeared a hydrophobic
301   liquid-solid interface, which was conducive to the enhancement of RRS.

302   (3) Enlargement of molecular volume

303        To our best knowledge, the RRS intensity is proportional to molecular volume or molecular
304   weight.[34] When AR combined with EV to form an ion-association complex, the molecular weight
305   increased. And this is the most important reason for RRS enhancement.

306   ## Conclusions

307        In summary, a new, simple, rapid and accurate method was presented for detecting allura red
308   based on triple-wavelength overlapping resonance Rayleigh scattering. Under the experimental
309   conditions, AR interacted with EV, a new RRS spectrum appeared with three peaks and the
310   scattering intensity increased dramatically. The analytical results showed that TWO-RRS method
311   was more sensitive than the commonly used single-wavelength RRS and would be impervious to
312   false signals arising by interferents. TWO-RRS method could be a great potential tool to enlarge
313   the applications of RRS.

314

315

316   ## Acknowledgements

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