Identification of tartrazine and sunset yellow by fluorescence spectroscopy combined with radial basis function neural network

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The fluorescence spectra of synthetic food dyes of sunset yellow and tartrazine are analyzed. The fluorescence peak wavelengths of sunset yellow and tartrazine are 576 and 569 nm, respectively, while the fluorescence spectra widths are 480–750 and 500–750 nm induced by ultraviolet light between 310–400 nm. The fluorescence spectra of sunset yellow overlap heavily with those of tartrazine, so it is difficult to distinguish them. Based on the principle of radial basis function neural network, a neural network is obtained from the training of the 14 groups of experimental data. The results show that the species of sunset yellow and tartrazine could be recognized accurately. This method has potential applications in other synthetic food dyes detection and food safety inspection.

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In recent years, many food safety issues such as sudan events occurred frequently. Due to the significance of food safety, it was regarded as the national public security item all around the world and has aroused widespread public concern. Food additives are commonly used in processed food-stuffs to improve appearance flavor, taste, color, texture, nutritive value, and conservation. Synthetic food dyes are used as the most important food additive. When compared to natural dyes, synthetic food dyes have shown many advantages such as bright luster, strong tinting strength, stability, and low price. However, synthetic food dyes are mainly the compounds of benzene or xanthene structure, which have some toxicity and could be converted to carcinogen in human body, and even cause damage. Therefore, the acceptable daily intake (ADI) for the varieties of synthetic food dyes must be strictly controlled. Strict supervision and effective inspection are required to ensure the food safety.

At present, a large number of analytical methods for synthetic food dyes have been proposed, such as ultraviolet (UV) spectrophotometry, high performance liquid chromatography, derivative adsorptive voltammetry[1−6], and so on. But few researches about the qualitative and quantitative characterization of synthetic food dyes by the fluorescence spectroscopy analysis are reported[7−13]. Because of its high accuracy and high sensitivity, fluorescence spectroscopy has been well applied in testing various substances in recent years. Due to the restriction of fluorescence spectra, the fluorescence spectra of synthetic food dyes need to be analyzed qualitatively. In practical applications, factors such as optical systematic error and stray light will affect the measurement precision. Synthetic food dyes contain abundant spectral information, thus it is not easy to form a parameterized mathematical model for the fluorescence spectra. The classification and recognition of synthetic food dyes are complicated nonlinear problems.

Radial basis function (RBF) neural network has been widely applied in many fields such as computer science, automation, pattern recognition, etc.[14,15] It can also be used to process experimental data to determine both the synthetic food dyes species and their relative concentrations with a much faster rate than, for example, non-negative least mean square estimation methods.

In this letter, several concentrations of sunset yellow and tartrazine are researched by fluorescence spectroscopy. We obtain the fluorescence spectral characteristics of the two kinds of synthetic food dyes. Combined with RBF neural network, the processing of the fluorescence spectroscopy is successful in recognizing the species of synthetic food dyes. This method combines the advantages of fluorescence spectroscopy and RBF neural network.

The experimental instrument is Sp-2558 multi-function spectral measurement system. The samples were sunset yellow and tartrazine standard solutions, each 0.5 ml (provided by the State Center for Standard Matter of China). Using solvent of ultrapure water, 12 solutions of different sunset yellow concentrations (1, 2, 5, 10, 20, 30, 40, 50, 60, 80, 100, 150 μg/ml) and 9 solutions of different tartrazine concentrations (1, 2, 5, 10, 30, 40, 50, 60, 100 μg/ml) were made.

The fluorescence spectra of sunset yellow and tartrazine at the concentration of 100 μg/ml were analyzed when induced by UV light between 310–400 nm. The three-dimensional (3D) equiangular and contour fluorescence spectra are shown in Fig. 1.

Table 1 shows the fluorescence spectral characteristics of sunset yellow and tartrazine. In the table, the best excitation wavelength is the excitation wavelength at which the relative fluorescence intensity reaches the maximum, and the fluorescence spectra width is the wavelength range of fluorescence spectra.

The similarity of both dyes on fluorescence peak wavelength and fluorescence spectra width could lead to fluorescence spectra overlapping seriously. The spectral res-

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olution obtained from traditional UV spectrophotometry or fluorescence spectroscopy is low, and the measurement error is large. RBF neural network, immune to most measurement noises, is able to compensate for the wavelength calibration error produced by the spectrometers.

RBF neural network is a network structure simulating the partial adjustment and mutually covering the receiving area or receptive field in human brain. It has been proved as a partial approaching network which approaches any continuous functions at any given accuracy. With the strong features of self-learning, self-organization, self-adaptive, high fault-tolerant capability, and associative memory, it has been widely applied in various fields and also achieved excellent results.

Figure 2[16] is the structural diagram of RBF neural network. R in the figure is the total number of input vector elements; P is the input vector; S^1 is the total number of hidden layer neurons; S^2 is the total number of linear layer neurons; $IW^{1,1}$ is the weight coefficient between input layer and hidden layer; $LW^{2,1}$ is the weight coefficient between hidden layer and linear layer; and Y is the output vector. The prototype of transfer function is radbas (n) $=\mathrm{e}^{-n^2}$. The RBF network is composed of three layers: input layer, hidden layer, and output layer. The input layer transmits signals to the hidden layer, in which the nodes are made up of the radiative function of image Gaussian function, and usually the output note is a simple linear function.

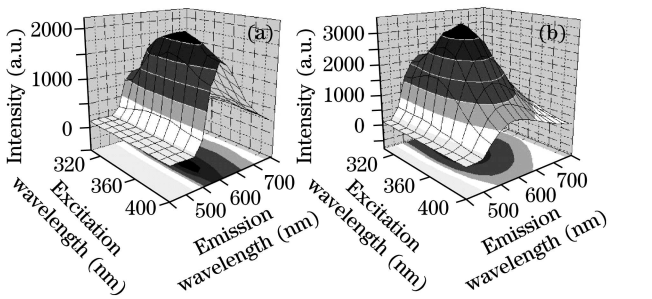

Fig. 1. 3D equiangular and contour fluorescence spectra of (a) sunset yellow and (b) tartrazine.

Table 1. Fluorescence Spectrum Characteristic of Sunset Yellow and Tartrazine

Sample	Sunset Yellow	Tartrazine
Best Excitation Wavelength (nm)	370	350
Fluorescence Peak (nm)	576	569
Fluorescence Spectra Width (nm)	480–750	500–750

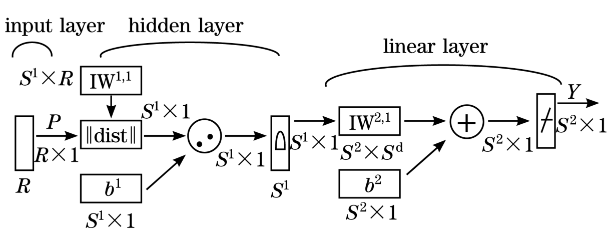

Fig. 2. Structure of RBF neural network.

Table 2. Sample Detection Results

Sunset Yellow	Sample (μg/ml)	20	30	40	80
	Result	1.0732	0.9618	0.9253	1.1128
Tartrazine	Sample (μg/ml)	2	40	50	
	Result	0.0806	0.0813	0.0765	

This work is based on the Matlab platform. The best excitation wavelengths of sunset yellow and tartrazine are 370 and 350 nm, respectively, so 360 nm was selected to irradiate the samples. Spectral data of different concentrations were divided, datum 8 of sunset yellow and datum 6 of tartrazine were selected as the training data, the other 7 groups of data as prediction data. Twenty groups of fluorescence intensity levels were selected from each spectrum, and one point every 10 nm was generated from the emission wavelength of 500–700 nm. Choosing 20 as the number of neurons of the input layer, 1 as the number the neurons of the output layer, we created an accurate network by a function called newrbe. The number of neurons of the hidden layer was selected automatically when the network RBF was created. The detection results were precise with the distribution density spread rate of RBF being 1.7.

In order to help the data analysis and network training, before running the RBF neural algorithm, we normalized the primitive data and delimited the output and input values of network range as [0, 1]. The normalization formula is

$$D_i' = \frac{D_i - D_{\min}}{D_{\max} - D_{\min}}, \quad (1)$$

where D_i is the non-normalized datum, $D_{\min}$ is the minimum datum of the training subgroup, $D_{\max}$ is the maximum datum of the training subgroup, and D_i' is the normalized datum.

The synthetic food dyes are classified with sunset yellow and tartrazine correspondingly representing 1 and 0. The sample detection results are listed in Table 2.

The reason why sunset yellow and tartrazine can emit fluorescence is discussed. There are some fluorescent chromophoric and auxochrome groups existing in the molecules, such as =C=O,>C=N−, benzene ring, naphthalene ring, –OH, $–SO_3Na$, etc. They are connected together through azo bond, leading to the formation of conjugated double-bond system. Electrons existing in it can easily absorb photons by the transition form of $\pi \rightarrow \pi^*$ and emit fluorescence. In addition, the conjugacy of molecules is enhanced when the substituents $–SO_3Na$ and –OH place at the para positions of naphthalene ring, and the molecules associates together through hydrogen bond. Figure 3 shows the molecular structures of sunset yellow and tartrazine.

The fluorescence spectral characteristics of sunset yellow and tartrazine standard solutions (Table 1) have shown that their fluorescence spectra overlaps heavily with each other. The RBF neural network program was able to recognize unknown samples with acceptable results (Table 2). When the result is about 1, sunset

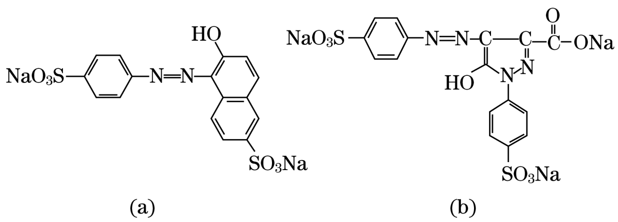

Fig. 3. Molecular structures of (a) sunset yellow and (b) tartrazine.

yellow would be recognized; when the result is about 0, tartrazine would be recognized.

In conclusion, the most common synthetic food dyes, sunset yellow and tartrazine, are researched through the analysis on fluorescence mechanism and detection on fluorescence spectroscopy. A RBF neural network is trained and constructed by using 14 groups of training sample spectral data. The unknown species of sunset yellow and tartrazine can be recognized accurately. This method can reduce the cost, shorten the analysis time, and improve the measurement accuracy and detection level greatly. It provides a new concept for food safety inspection especially when food safety becomes more and more important.

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