Feature Extraction of Fourier Infrared Signals from Pyrolysis Products based on ZCA and PSO

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***Abstract*—Fourier transform infrared spectroscopy (FTIR) can provide abundant information for different organic functional groups and chemical bonds, and is therefore widely used in molecular structure analysis, qualitative analysis of mixtures, and quantitative analysis. However, it is difficult to decompose and extract the highly correlated component spectra from complex mixed spectrum, and existing methods for FTIR qualitative analysis has poor recognition effect. For this reason, a spectroscopic analysis model based on zero-phase component analysis (ZCA) whitening and particle swarm optimization (PSO) is proposed in this paper. The model uses wavelet threshold filter to de-noise the mixed spectrum, and uses ZCA whitening to remove the correlation of the component spectra. Finally, PSO is used to solve the component concentration. In order to verify the superiority of the ZCA whitening-based spectral analysis model, simulation studies are carried out on signals made up by twenty kinds of component spectra. The simulation results show that the model not only achieves effective qualitative analysis and quantitative analysis of the main components of the mixed spectrum, but also has better performance than the current mainstream spectrum analysis model.**

Keywords—Fourier transform infrared spectroscopy, Pyrolysis products, Particle swarm optimization (PSO), Wavelet threshold filter, Zero-phase component analysis (ZCA) whitening

I. INTRODUCTION

Fourier transform infrared spectroscopy (FTIR) can provide a wealth of information on different organic functional groups and chemical bonds. At present, scholars use Fourier transform infrared spectroscopy to conduct qualitative and quantitative analysis of substances. There are mainly peak matching methods, correlation analysis methods, partial least squares methods, and principal component analysis algorithms. Among them, the peak matching method is a hot topic for domestic scholars. The main analysis method of the Fourier transform infrared spectrum of the solution products. However, due to the large number and complex composition of pyrolysis products of domestic waste, the above four methods cannot meet the requirements of qualitative and quantitative analysis of Fourier transform infrared spectrum of pyrolysis products of domestic waste.

Thesis [3] used FTIR to study the infrared absorption spectral characteristics of major substances in industrial exhaust gas[3]. Thesis [4] analyzes the characteristic peak wave numbers of the main six gas products, and qualitatively judges each gas based on the characteristic peak absorbance of the infrared absorption spectrum of gas products[4]. The paper [5] uses signal analysis theory to obtain the correlation between pure components and mixed components through correlation processing[5]. In view of the large number of complex products of domestic waste pyrolysis products, this paper proposes a spectral analysis model based on ZCA albinism and PSO concentration. This model does not require a priori information of the main components of the mixed spectrum and can be used to accurately identify materials with similar structures. Besides, original multivariate variables problem transforming to a single variable problem due to the whitening process, the computational cost has been greatly reduced.

The rest of the paper is organized as follows. We introduce the spectral data preprocessing methods, which mainly introduces the wavelet threshold to Noise principle and method in Section 2. A spectral analysis model based on ZCA whitening and correlation analysis are given in Section 3. The ZCA whitening-based spectrum model and the comparison with the existing infrared spectrum analysis models are presented in Section 4. Conclusions are given in Section 5.

II. DATA CLUSTERING

A. The standard library construction

The Fourier transform infrared spectrum of the pyrolysis products collected in this paper is the Fourier transform infrared spectrum of the pyrolysis gas at each moment in the pyrolysis process. The spectrum reflects the generation rate information of the pyrolysis gas.

The main gas products of pyrolysis of domestic garbage are CO_2, CO, CH_4, C_2H_4, C_2H_6, C_3H_6, C_3H_8, C_4H_{10}, H_2 and tar [11].

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Among them, tar is a complex mixture of highly aromatic hydrocarbons. Its main components are polycyclic, fused ring compounds with or without side chains, and heterocyclic compounds containing oxygen, sulfur, and nitrogen. Aliphatic, naphthenic and unsaturated hydrocarbons.

In this paper, 29 standard infrared spectra of gases downloaded from the NIST chemical library are used for simulation experiments. The 29 spectra include 1,2-butadiene, 1,3-butadiene, 1-butene, 1-butyne, 1-propene, 2-methyl, 2-butene, cis, 2-butene, trans, 2-butene, 2-butyne, ammonia, acetaldehyde, benzene, butane, carbon dioxide, carbon monoxide, cyclopropane, ethane, ethanol, ethylene, formaldehyde, hydrogen sulfide, methane, methanol, phenol infrared spectra of propane, 2-methyl, propane, propylene, toluene, and water vapor. The collection of the 29 standard spectra is referred to as the standard library in the follow-up of this paper.

Theoretically, the more spectra the standard spectral library is constructed, the better the model's multi-infrared spectral analysis performance. It should be noted that, because there is no infrared spectrum for hydrogen, the Fourier transform infrared spectrum analysis technology cannot analyze the hydrogen in the pyrolysis gas.

B. The spectrum pretreatment

Due to instrument noise, light source intensity, or data processing requirements, the original spectrum measured by a Fourier transform infrared spectrometer cannot be used directly for analysis. The original Fourier transform infrared spectrum needs simple data processing before it can be applied to the spectral analysis. The general methods of spectral preprocessing include: baseline correction, spectral normalization, and spectral smoothing.

1) Baseline correction

In the process of measuring the spectrum using a Fourier transform infrared spectrometer, due to factors such as the light source intensity, response of the detector, and fluctuations in light intensity, the basis of the infrared spectrum often shifts, bends, and the tilt causes the baseline to not reach the desired horizontal state. At this time, the baseline needs to be corrected first.

There are two methods for baseline correction: automatic and manual. This design uses the automatic baseline correction that comes with OMNIC software. The eleventh minute pyrolysis products of the pyrolysis experiment of the municipal solid waste (hereinafter referred to as 50 MSW) at a heating rate of 50 K/min The spectrum is taken as an example. The spectra before and after baseline correction are shown in Fig. 1 and Fig. 2.

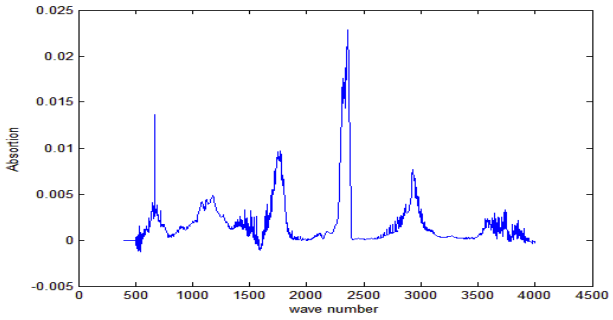

Fig. 1. Infrared spectrum of 50 MSW pyrolysis products at 11 minutes without baseline correction

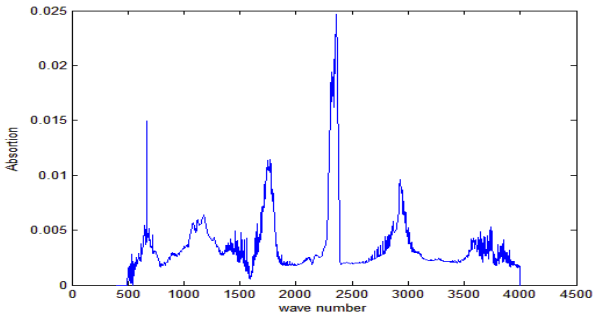

Fig. 2. Infrared spectrum of 50 MSW pyrolysis products at 11 minutes with baseline correction

2) Spectrum normalization

The spectrum normalization process includes ordinate normalization and abscissa normalization. The standard spectrum obtained from the spectral library is not a normalized infrared spectrum. The un-normalized spectrum will make subsequent spectral analysis more complicated. Therefore, the standard spectrum needs to be normalized so that the maximum peak value of the spectrum is 1. The ordinate normalization is described as follow:

$$T = \frac{T}{\max(T_i)} \tag{1}$$

where T is the transmittance, that is, the ratio of the intensity of the emitted light and the intensity of the incident light; T_i is the transmittance of the wavenumber.

Because the resolution of the pyrolysis product spectrum and the standard spectrum downloaded from the spectral library do not match the resolution and wavenumber domain, the abscissa of the spectrum needs to be standardized to make the resolution of the analysis spectrum consistent with the wavenumber domain. We use the interpl() function of MATLAB to perform linear difference processing on the spectrum:

$$T_j = T_{j^-} + \frac{j - j^-}{j^+ - j^-} \times (T_{j^+} - T_{j^-}) \tag{2}$$

where j is the expected wavenumber, and j^+ as well as j^- represent two adjacent wavenumber to the wavenumber j. The resolution of the processed spectrum is $1cm^{-1}$, and the wavenumber domain is $600cm^{-1} \sim 3764cm^{-1}$.

3) Spectrum smoothing

In the actual spectrum measurement, due to the influence of uncertain factors such as instrument and environment, the measured spectrum always adds extra noise. The smoothing technology averages the original infrared spectrum to a certain degree, which can reduce the high-frequency noise. In this design, we use wavelet threshold de-noising to smooth the original spectrum.

According to different threshold selection criteria, threshold function selection methods, and threshold processing changes with noise levels, wavelet threshold de-noising is divided into several different methods. Firstly, we choose the heuristic threshold principle as the threshold selection criterion. Heuristic threshold de-noising is a combination of the principle of unbiased likelihood estimation and the principle of fixed threshold. When the signal-to-noise ratio is small, the unbiased likelihood estimation principle is used; when the signal-to-noise ratio is large, the fixed threshold principle is used. Besides, we choose the soft threshold as the threshold function selection method. For wavelet coefficients with modulus less than 3, soft threshold de-noising is all set to zero; for wavelet coefficients with modulus less than -3, soft threshold de-noising is added by 3; for wavelet coefficients with modulus greater than 3, soft threshold de-noising method is selected. The wavelet coefficient of the processed signal is relatively smooth, which avoids the influence of local jitter caused by hard threshold de-noising. And we select the threshold processing to adjust based on the noise level estimation of the first wavelet decomposition. Because the change of white noise in the frequency domain is generally more severe than that of the signal, it can be considered that the first layer of wavelet coefficients mainly saves the information of the noise. Adjusting the threshold based on the noise level estimation of the first layer of wavelet decomposition can be compared to the ideal de-noising effect.

We use coif5 wavelet to perform 6-layer decomposition on the spectral signal of the pyrolysis product, and use the MATLAB wden() function to perform wavelet threshold de-noising on the signal. It is worth noting that in the process of de-noising the pyrolysis product signal, the minimum value of the de-noised signal may be less than 0. At this time, the minimum value of the de-noised spectrum needs to be subtracted to complete the baseline correction. The de-noising process is briefly depicted in Fig.3.

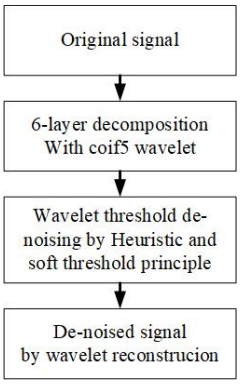

Fig. 3. Process of spectrum smoothing by wavelet de-noising

Taking the temperature spectrum of 50K / min pyrolysis experiment at the eleventh minute as an example, the spectra of the pyrolysis products are shown in Figs. 4 and 5 respectively before and after de-noising.

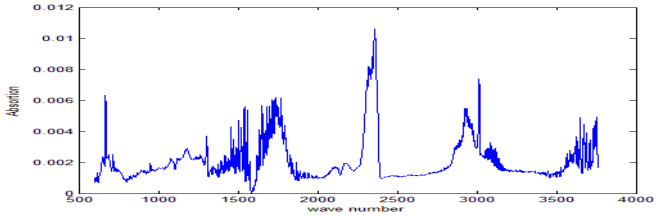

Fig. 4. 50 MSW 11th minute pyrolysis product spectrum

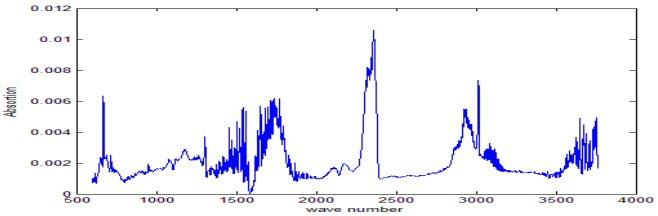

Fig. 5. De-noised spectrum of pyrolysis product at 50 MSW at 11th minute

III. SPECTRAL ANALYSIS MODEL BASED ON ZCA ALBINO AND PSO CONCENTRATION

A. Fundamental analysis of Fourier transform infrared spectroscopy

The basis of Fourier transform infrared spectroscopy theoretical analysis is Lambert's law. Lambert's law is the basic law of spectrophotometry, which is used to describe the strength of a substance to absorb light at a certain wavelength and the concentration of light-absorbing substances and their liquid layers. The definition is described as follows. When a parallel monochromatic light passes through a uniform non-scattering sample, the absorbance of the sample is proportional to the concentration and thickness of the sample [13]. Its mathematical expression is:

$$A = \lg(1/T) = Kbc \tag{3}$$

In (3), A is the absorbance; K is the spectral absorption coefficient, which is related to the nature of the absorbing substance and the wavelength of the incident light; c is the concentration of light-absorbing substance; b is the thickness of the absorbing layer. In monochromatic light, Lambert's law is expressed in the wavenumber domain as follows:

$$T(v) = e^{-K(v)bc} \tag{4}$$

When monochromatic light passes through a multi-component material, Lambert's law can be expressed as:

$$T(v) = e^{-\sum_{i}^{n} K_i(v)bc_i} \tag{5}$$

$$\mathrm{A} = \lg(1/\mathrm{T}(v)) = \sum_{i}^{\mathrm{n}} \mathrm{K}_i(v)bc_i \tag{6}$$

Based on equation (5-6), the matrix expression of Lambert's theorem is as follows:

$$A_{m\times 1} = K_{m\times n} C_{n\times 1} + e_{m\times 1} \tag{7}$$

where $A_{m\times1}$ is the light absorption concentration of the mixed spectrum, $K_{m\times n}$ is the absorbance matrix of the component spectrum, $C_{n\times1}$ is a concentration matrix, and $e_{m\times1}$ is error.

B. ZCA whitening

The Fourier transform infrared spectrum theorem analysis technique based on Lambert's theorem essentially solves the absorbance matrix and the concentration matrix. The methods to solve (7), which is an underdetermined matrix equation, include orthogonal matching pursuit method. However, as substances with same functional groups and chemical bonds are strongly similar in Fourier infrared spectroscopy, the methods above are hard to get accurate results.

In this paper, an information lossless method, ZCA is selected as the whitening-based spectral analysis method. The fundamental principle is the same as the orthogonal matching pursuit method. The core idea is to use the irrelevance of orthogonalized spectra to solve the underdetermined matrix equation. The model first performs ZCA whitening on the spectrum of the standard spectrum library after data preprocessing and obtains the whitening matrix to remove the correlation in the spectrum.

Then, the mixed spectrum is processed using the whitening matrix; the standardized library after whitening and the whitened correlation coefficients of the mixed spectra are used to determine the main gas components of the pyrolysis products.

C. Concentration calculation by PSO

Because the ratio of the correlation coefficients of the main components of the mixed spectrum is approximately equal to the ratio of their concentrations, after determining the main components of the mixed spectrum and obtaining the correlation coefficients of the main components and the simulated spectrum, the main component spectra can be combined using the correlation coefficients into a mixed base spectrum. Solving the problem of the concentration of the main components of the mixed spectrum can be transformed into the problem of solving α, the ratio of the absorbance of the mixed spectrum composed of only the main components to the absorbance of the mixed base spectrum, where α is equal to the ratio of the concentration of the main components of the mixed spectrum to the correlation coefficient. Its mathematical model is as follows:

$$A = \lg(1/T) = Kbc \tag{8}$$

$$A_{m\times1} = K_{m\times n} C_{n\times1} \tag{9}$$

$$C_{n\times1} = \alpha Corr_{n\times1} \tag{10}$$

$$A_{m\times1} = \alpha A_{base_{m\times1}} \tag{11}$$

where $K_{m\times n}$ is the main component spectral absorbance matrix, $Corr_{n\times1}$ is the correlation coefficient matrix of the main components of the whitening spectrum and the whitening simulation spectrum, $C_{n\times1}$ is the concentration matrix of the main components of the mixed spectrum, $A_{base_{m\times1}}$ is the mixed-base spectrum, and $A_{m\times1}$ is the mixed spectrum composed of main components.

To solve α, a novel object function that aims to minimize the correlation between the ZCA whitened de-noised spectrum A_D^Z and the rest ZCA whitened spectrum A_{Ri}^Z except main components is proposed. Since the object function is not derivable to α, PSO is applied to get the solution. Compared with other heuristic algorithms, PSO has less parameters resulting in easy implementation and faster calculation. The PSO algorithm is described as follow:

$$v_i^k = (w\cdot d)v_i^{k-1} + c_1 r_1(pbst_i - \alpha_i^{k-1}) + c_2 r_2(gbst - \alpha_i^{k-1}) \tag{12}$$

$$\alpha_i^k = v_i^k + \alpha_i^{k-1} \tag{13}$$

$$\alpha = \arg\min_{\alpha} \sum_i \mathrm{CORR}(A_D^Z, A_{Ri}^Z) \tag{14}$$

where v_i^k represents the speed of the ith particle in the kth, the number of particles equals to 50, the number of max iteration equals to 1000; w is the initial velocity inertia and equals to 0.9; d is the attenuation coefficient and equals to 0.1; c_1 and c_2 are learning factors and both equal to 0.05; r_1 and r_2 are random numbers defined between 0 and 1; $pbst_i$ is the historical optimal fitness position of the ith particle; $gbst$ is the current global optimal position of the group; CORR(,) is the spearman correlation function. Besides, the threshold of searching termination sets to 1e-5. The process of complete spectrum analysis is depicted as Fig. 6.

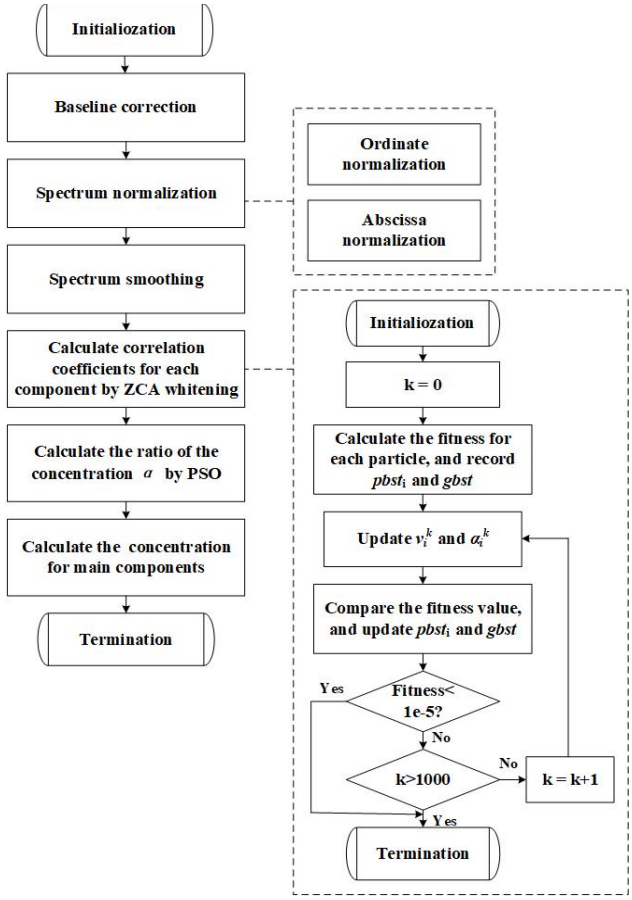

Fig. 6. Complete spectrum analysis process

IV. MODEL SIMULATION AND RESULT ANALYSIS

A. Simulation steps

In this paper, simulation experiments are performed on simulated spectra constructed from standard spectral library spectra to verify the validity of the spectral analysis model based on ZCA albinism and PSO concentration. The steps of model simulation include the construction of simulation spectrum, the spectral ZCA whitening, and the use of particle swarm algorithm to solve the component spectral concentration.

1) Construction of simulated spectrum

In order to verify the validity of the ZCA whitening-based spectral analysis model for analyzing complex mixed spectra, the mixed spectra constructed in the simulation consists of the 29 spectra, and the concentration settings are shown in Table I.

TABLE I. MIXED SPECTRAL COMPOSITION AND CONCENTRATIO

Spectrum number	Component name	Concentration
14	Carbon dioxide	0.8
15	Carbon monoxide	0.7
22	Methane	0.62
17	Ethane	0.57
19	Ethylene	0.5
26	Propane	0.43
27	Acrylic	0.3
13	Butane	0.25
23	Methanol	0.11
1	1,2-butadiene	0.08
18	Ethanol	0.05
11	Acetaldehyde	0.023
20	Formaldehyde	0.02
29	Steam	0.013
2	1,3-butadiene	0.008
8	2-butene	0.006
9	2-butyne	0.001
10	Ammonia	0.0007
24	Phenol	0.0003
25	Propane, 2-methyl	0.00008

Fourier infrared spectroscopy reflects information about the functional groups and chemical bonds of substances. Substances with the same functional groups and chemical bonds will have larger peaks in the wavenumber domain corresponding to the functional group. The overlapping of the peaks brings difficulties to the analysis of the infrared spectrum. In order to verify the validity of the model, we specifically set a set of isomers in the composition of the mixed spectrum-1,2-butadiene and 1,3-butadiene, of which the concentration of 1,2-butadiene is 1,3 -butadiene 10 times. The isomer setting is used to verify the validity of the ZCA albino-based spectral analysis model in extracting the correct isomer material from the mixed spectrum. Figs. 7 and 8 show the spectra of 1,2-butadiene and 1,3-butadiene, respectively.

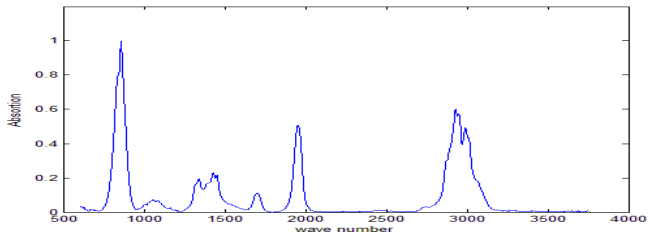

Fig. 7. 1,2-butadiene spectrum

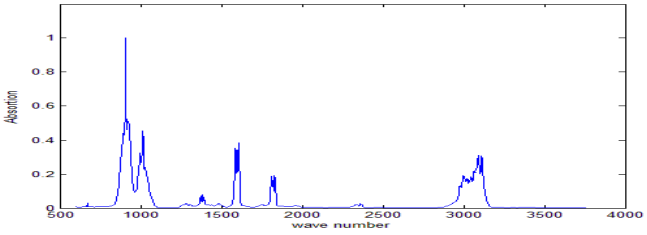

Fig. 8. 1,3-butadiene spectrum

Acetaldehyde and formaldehyde have the same functional group aldehyde and methyl groups, but the number of methyl groups is different. In the composition of the mixed spectrum, the concentrations of Acetaldehyde and formaldehyde are set similarly. The purpose is to test the effectiveness of the ZCA whitening-based spectral analysis model that can identify substances with the same functional groups and similar concentrations. After that, add 35dB Gaussian white noise to the mixed spectrum to obtain the final simulated spectrum.

2) Main components of mixed spectrum

The spectral analysis model based on ZCA whitening and PSO concentration proposed in this paper uses ZCA whitening to remove the correlation between the spectra to obtain the whitening standard library spectrum, and uses the whitening matrix to process the simulated spectrum to obtain the whitened simulated spectrum. The correlation coefficient was obtained from the spectrum of the whitened standard library.

TABLE II. CORRELATION COEFFICIENT BETWEEN ZCA WHITENING STANDARD SPECTRUM AND WHITENING SIMULATION SPECTRUM

Spectrum number	Correlation coefficient	Spectrum number	Correlation coefficient
14	0.055754	18	0.003598
15	0.048789	11	0.001686
22	0.042336	20	0.001196
17	0.039383	4	0.001069
19	0.033537	8	0.000936
26	0.029370	29	0.000915
27	0.020043	9	0.000730
13	0.016997	2	0.000532
23	0.007339	7	0.000375
1	0.005556	16	0.000232
10	0.000187	5	-0.000100
25	0.000101	3	-0.000122
21	0.000072	12	-0.000231
24	0.000031	6	-0.000234
28	-0.000003		

As can be seen from Table Ⅱ, the ZCA whitening-based spectral analysis model can effectively identify the main components in the mixed spectrum, and the concentration ordering of the first 13 component spectra of the model is completely correct. However, when the component spectral concentration is low, the component spectrum of the model will be wrong. The aggregation absorbance of the rest 7 components is depicted in Fig. 9 . The amplitude of the signal in Fig.9 can be covered by noise in majority wavenumbers. Hence, we believe the reason of the incorrect order of low concentration components is that the noise has a greater effect on the spectrum of the lower concentration components, and the characteristic information may be lost in the spectrum of the lower concentration components while removing the noise. For this reason, in the subsequent process of determining the concentration of the mixed spectrum components, only the concentrations of the main constituents of the mixed spectrum are solved.

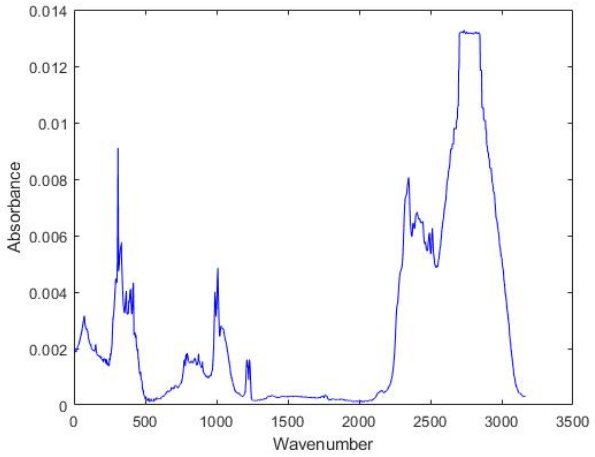

Fig. 9. Aggregation absorbance of the disorder components

The determination of the main components of the mixed spectrum is the key to accurate analysis of the Fourier transform infrared spectrum. The determination of the main components is not only affected by the spectral concentration of the components, but also by the de-noising algorithm. After many experiments, using the coif5 wavelet to perform 6-layer decomposition of the spectrum is a better wavelet threshold de-noising combination.

In Table Ⅱ, the correlation coefficient of 1,2-butadiene with higher component concentration is much larger than the correlation coefficient of 1,3-butadiene with lower component concentration. This result shows that the spectral analysis model based on ZCA whitening has an accurate advantage of distinguishing isomers.

3) Solving the main component concentration of mixed spectrum

After determining the main components of the mixed spectrum and obtaining the correlation coefficients of the main components and the simulated spectrum, PSO is applied to solve the ratio of the concentration of the main components. And the concentration of main components is solved by equation (10).

The main component concentration results and errors solved by the PSO are shown in Table Ⅲ. It can be seen that errors are so small that the superior performance of the proposed algorithm has been proved

TABLE III. CALCULATION RESULT OF MAIN COMPONENT CONCENTRATION

Spectrum number	Component name	True concentration	Calculated concentration	Error (%)
14	Carbon dioxide	0.8	0.7981	0.24
15	Carbon monoxide	0.7	0.6952	0.68
22	Methane	0.62	0.6053	2.37
17	Ethane	0.57	0.5569	2.29
19	Ethylene	0.5	0.4862	2.76
26	Propane	0.43	0.4229	1.64
27	Acrylic	0.3	0.2778	7.41
13	Butane	0.25	0.2421	3.16
23	Methanol	0.11	0.1059	3.71
1	1,2-butadiene	0.08	0.0784	1.99
18	Ethanol	0.05	0.0512	-2.38
11	Acetaldehyde	0.023	0.0227	1.52

B. Simulation steps

This section takes the simulated spectrum constructed in Section A as the experimental object, and compares the performance of qualitative analysis with quantitative analysis of the existing infrared spectrum analysis model and the spectral analysis model based on ZCA albinism and PSO concentration solution proposed in this paper.

The purpose of qualitative spectral analysis is to identify the main components in the mixed spectrum, and to whiten the standard spectrum, which is helpful for the model to correctly identify the main components in the mixed spectrum. At present, the qualitative analysis method of spectrum is principal component regression analysis, which uses PCA to remove the correlation between spectra. However, PCA only uses the information with the largest difference to whiten the data, which causes information loss to the whitened data.

Compared with the ZCA whitening used in the spectral analysis model based on ZCA whitening and PSO concentration proposed in this paper, the spectrum of PCA will be reduced and the information will be lost after whitening. Table Ⅳ compares the qualitative analysis performance of models that do not use whitening, PCA-based whitening and ZCA-based whitening.

TABLE IV. COMPARISON OF QUALITATIVE ANALYSIS PERFORMANCE OF MODELS BASED ON DIFFERENT WHITENING TREATMENTS

Whitening method	Recall
No whitening	0
PCA	93.04%
ZCA	99.35%

It can be seen from Table Ⅳ that, for complex mixed spectra, accurate qualitative analysis of the mixed spectrum cannot be achieved without whitening the spectrum. Although the recall rate of the spectral analysis model based on ZCA whitening and the spectral analysis model based on PCA whitening is only 6.31%, the recall rate of the spectral analysis model based on ZCA whitening is close to 100%, which indicates that the spectral analysis model based on ZCA whitening is effective for small The identification performance of the components far exceeds the spectral analysis model based on PCA whitening. Therefore, the spectral analysis model based on ZCA albinism and PSO concentration solution proposed in this paper is stronger than the existing spectral analysis model in terms of spectral qualitative analysis capabilities.

Besides, to verify the effectiveness of the object function that aims to minimize the correlation between A_D^Z and A_{Ri}^Z, least squares method that aims to minimize the loss information between A_D^Z and the reconstruction spectrum composed of main components. The comparison of RMSEC is shown in TABLE Ⅴ. It can be seen that the performance of the proposed method is far better than the least square method.

TABLE V. COMPARISON OF QUANTITATIVE ANALYSIS PERFORMANCE OF MODELS BASED ON DIFFERENT ALGORITHMS

Algorithm	**Number of main components**	**RMSEC**
Least squares	13	0.209876
Proposed method	13	0.008552

V. CONCLUSION

In this paper, a spectral analysis model based on ZCA whitening is proposed. This model includes wavelet threshold de-noising and standard spectral library ZCA whitening, and uses the PSO to solve the concentration of the main components of the mixed spectrum. The proposed method transforms the original multivariate variables problem to a single variable problem that simplifies the calculation. In order to verify the effectiveness of the proposed model, a mixed spectrum is constructed using 20 gas component spectra for simulation experiments. Simulation results show that the proposed model can accurately identify the main components of the mixed spectrum. The results of the model calculation of the main component concentrations are also generally better than other methods.

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