Study on the Determination Method of Atomizing Agent Content in Heat-not-burn Reconstituted Tobacco by Microwave Technology

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Abstract. This paper presents an online determination system for atomizer content in heat-not-burn reconstituted tobacco established by a dual-band microwave sensor independently developed. Employing Ridge regression analysis, collinear variables were effectively eliminated, and combined with gas chromatography, a robust determination model for glycerol content in reconstituted tobacco was successfully established. In addition, the repeatability and stability of the developed determination system were rigorously evaluated. The evaluation results indicate that: (1) The model exhibits a high accuracy with a correlation coefficient of 0.982; the average error is merely 0.33%, and the standard deviation is 0.41%. (2)The system demonstrates excellent measurement precision and stability, effectively overcoming the drift problems of measurement errors over time. This system not only meets quality control needs in reconstituted tobacco production and improves product quality, but also significantly enhances detection efficiency. It is of extremely important significance for promoting the improvement of reconstituted tobacco efficiency and realizing accurate control of product quality.

Keywords: Microwave determination technology; Heat-Not-Burn reconstituted tobacco; glycerol; ridge regression.

1. Introduction

Heat-not-burn reconstituted tobacco are an innovative category of tobacco product and a form of reconstituted tobacco leaves. This approach facilitates the release of flavor substances and nicotine through heating rather than combustion, thereby reducing the noxious substances typically produced during the incineration process associated with conventional cigarettes. Consequently, this has become a novel frontier for research and development in the industry [1-3]. Due to the low heating temperature for heat-not-burn reconstituted tobacco(not exceeding 350℃), the aerosol emission is also lower than combusted cigarettes [4]. To meet the smoking needs of consumers and increase the smoke release amount in the heat-not-burn reconstituted tobacco, tobacco atomizing agents (such as 1,2 - propylene glycol and glycerol) are usually added [5,6]. Glycerol and other atomizing agents in heat-not-burn reconstituted tobacco can not only increase the hygroscopicity and water retention capacity of the sheets and improve the smoking taste [7], but aslo enhance the thermal conductivity of the sheets, boost the heating efficiency, and promote the volatilization of smoke constituents [8]. Therefore, its content plays a crucial role in the generation mechanism and aroma release of heated tobacco products. Accurate measurement of the atomizing agent content is of great significance. Currently, the detection methods for atomizing agents such as 1,2-propanediol and glycerol primarily involve gas chromatography, which is complex to operate and time-consuming, and it is destructive to the samples being tested. This does not meet the requirements for rapid and non-destructive testing in actual production processes, thereby limiting the production efficiency of companies. In exploring novel methodologies, Xing Zishuo [9] established a determination model for the moisture content of heat-not-burn reconstituted tobacco with ridge regression method whitch was based on a microwave moisture density meter, achieving a correlation coefficient of 0.95. This has realized the application of the microwave method in the determination of related substance contents in heat-not-burn reconstituted tobacco, but it did not investigate the determination of atomizing agents like glycerol. Consequently, this paper designs an online determination device for the atomizing agent content of heat-not-burn reconstituted tobacco based on microwave technology. By designing a dual-frequency microwave sensor, extracting the characteristic response parameters of sheet atomizing agents, and combining chemometrics, rapid and non-destructive detection of glycerol content is achieved, effectively speeding up production efficiency and improving product quality, and enhancing the automation level of sheet production. This has important practical significance for promoting technological progress and productivity in the tobacco industry.

2. Principle of microwave detection

Microwave perturbation method was used to determine the content of atomizing agent content in heat-not-burn reconstituted tobacco. In this method, the dielectric material s with special dielectric properties is placed into the microwave resonator T. If the volume of s is relatively small, a slight volume perturbation is generated relative to the cavity space. The perturbation causes changes of resonant frequency F and resonant bandwidth Δ f, and further leads to the change in the quality factor Q of the resonator. By precisely measuring the changes in f and Q, the dielectric constant of the dielectric material can be calculated [10] .

In general, the dielectric constant of a dielectric material is characterized in a complex form as shown in Formula (1) .

$$\varepsilon = \varepsilon' + j\varepsilon'' \tag{1}$$

In the formula, ε ' is the real part of the dielectric constant, which indicates the energy storage capacity and can cause changes in the frequency and phase of the electromagnetic wave passing through. ε ” is the imaginary part, which characterizes the energy dissipation capacity and can cause the attenuation of the energy of passing electromagnetic wave [11] .

Taking the cylindrical microwave resonator as an example, there is a mathematical relationship ,as shown in Equation (2), between the resonant frequency f, the quality factor Q and the dielectric constant of the dielectric material:

$$\begin{cases} \frac{f_s - f_0}{f_0} = -2 \cdot \left(\varepsilon' - 1\right) \cdot \frac{V_s}{V_0} \\ \frac{1}{\Delta Q} = \frac{1}{Q_s} - \frac{1}{Q_0} = 4 \cdot \varepsilon'' \cdot \frac{V_s}{V_0} \end{cases} \tag{2}$$

In the formula: fs and f0 represent the resonant frequencies of the microwave resonator with and without sample load, respectively; Vs and V0 are the volumes of the resonator and the dielectric sample, respectively; Qs and Q0 are the quality factors of the microwave resonator with and without sample load. The measurement method of the quality factor Q primarily involves measuring the resonant frequency f and the bandwidth Δ f, as follows:

$$Q = f/2\Delta f \tag{3}$$

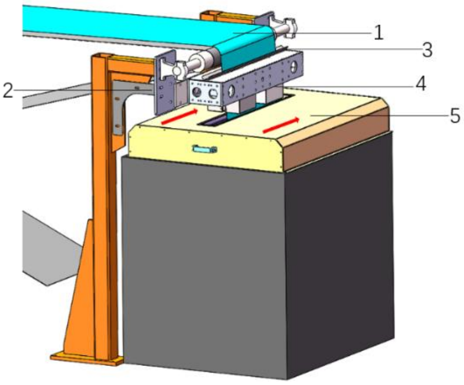
The formula (4) can be obtained by substituting formula (3) into formula (2) . From this, the final measurement parameters of dielectric materials by microwave perturbation method are the resonant frequency f and the resonant bandwidth Δf of the microwave sensor.

$$\begin{cases} \varepsilon' = -2 \cdot \frac{f_s - f_0}{f_0} \cdot \frac{V_0}{V_s} + 1 \\ \varepsilon'' = \frac{1}{4} \cdot \frac{V_0}{V_s} \cdot \left(\frac{\Delta f_s}{f_s} - \frac{\Delta f_0}{f_0}\right) \end{cases} \tag{4}$$

Generally, in heat-not-burn reconstituted tobacco, the principal components of atomizing agents are 1,2-propanediol and glycerol, among which the content of glycerol is relatively high. According to the "Dielectric Constant Table", it can be known that at 25°C, the dielectric constants of 1,2-propanediol and glycerol are 32 and 42.5 respectively, and the dielectric constants of most other absolutely dry substances are 1 to 5. This significant difference provides a theoretical possibility for the microwave perturbation method to determination the dielectric constant of the atomizing agent [12]. Based on the principle of microwave resonant cavity perturbation method, the study measures the dielectric constant parameters of the atomizing agent of heat-not-burn reconstituted tobacco in the microwave resonant field, and combines standard methods such as gas chromatography to measure the atomizing agent content. By establishing a mathematical model of "dielectric constant parameter - atomizing agent content", it is possible to achieve the determination of the atomizing agent content in heat-not-burn reconstituted tobacco.

3. Detection System

In order to achieve online measuring of heat-not-burn reconstituted tobacco, this study developed an online measuring system for sheet atomizing agents based on the microwave perturbation method. The system mainly consists of a microwave unit and a control unit. The microwave unit includes a dual-band microwave sensor, a microwave signal source, a demodulation module, and a data acquisition module. In addition, sample data are collected and uploaded at a frequency of 15 milliseconds to ensure real-time measuring of the online reconstituted tobacco production process. By transferring the packeted sample data to the control unit and processing them with related algorithms, the system achieves accurate determination of the atomizer content.



1.reconstituted tobacco Production Line 2. Auxiliary Structure 3. Guide Plate 4. Microwave Determination Device 5. Cutter protection cover

Fig. 1 structure diagram of on-line detecting system for heating non-burning film atomizer

3.1 Design of dual-band microwave sensor

Because the relaxation of water molecules is especially obvious in the microwave frequency range from 2.0 GHz to 4.0 GHz, the microwave equipment widely used in tobacco moisture measuring is mainly concentrated in this frequency range, which makes moisture detection technology quite mature in the application, and has high accuracy. However, when using microwave signals within the same frequency band to determinate the atomizing agent content in heat-not-burn reconstituted tobacco, the interactions between 1,2-propanediol, glycerol, and moisture will lead to a decrease in measuring accuracy, which may even render effective measurement impossible. To address this issue, a microwave sensors with two different resonant frequencies were employed to measure the glycerol content in reconstituted tobacco. The lower frequency is about 1 GHz, which can effectively reduce the response of water molecules in sheet detection and enhance the microwave response of glycerol, while the higher frequency is about 4 GHz. By accurately measuring the water content, the interference of water on the measuring of glycerol content is eliminated. In addition, the two resonant can provide different frequencies and phases of alternating electric field, as well as independent resonant bandwidths, a total of 6 independent parameters and multiple cross-parameters. Through the ridge parameter regression analysis of the above parameters, parameters with strong correlation are screened out, and the determination model for glycerol content in sheets is established.

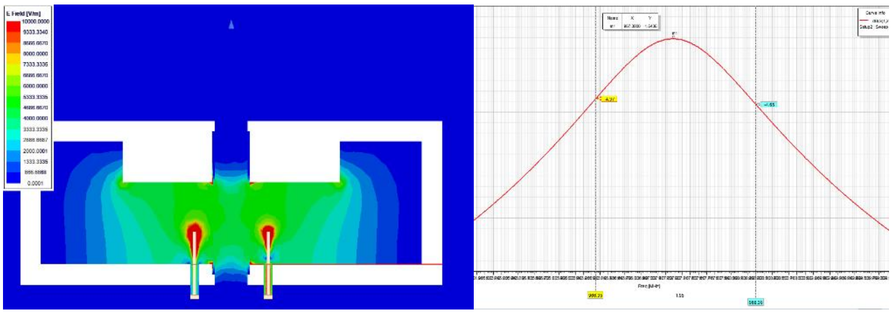


Fig. 2 1GHz simulation results of electric field distribution and S12

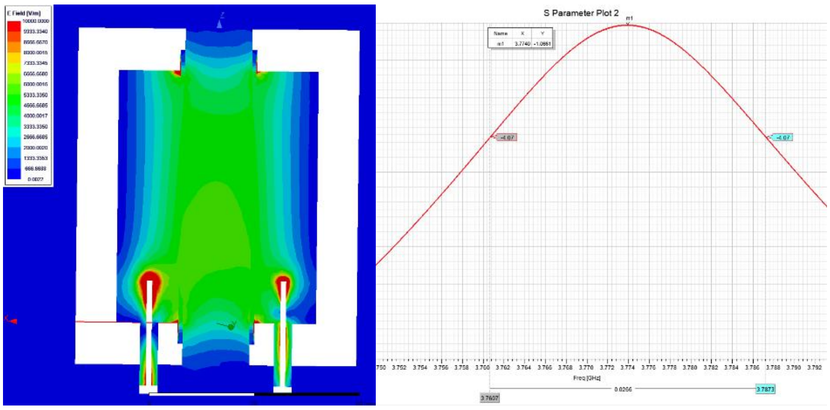


Fig. 3 4GHz simulation results of electric field distribution and S12

3.2 Ridge regression analysis

Based on the research and design of dual-band microwave sensors, a number of related variables have been formed, and there may be multicollinearity among these independent variables. Therefore, ridge regression is introduced in the research to achieve the effect of eliminating the influence of collinearity. Ridge regression is an improved least squares method. In essence, by combining the penalty function with the L2 regularization term, more practical and reliable regression coefficients are obtained at the cost of losing some information and reducing accuracy.

$$\widehat{\beta}^{ridge} = argmin\left\{\sum_{i=1}^{N}\left(y_i - \beta_0 - \sum_{j=1}^{p} x_{ij}\beta_j\right)^2 + \lambda\sum_{j=1}^{p}\beta_j^2\right\} \quad （5）$$

Formula (5) represents the loss function of the ridge regression model, where: y is the output vector, x is the p-dimensional feature vector, β is the coefficient vector, and $\lambda\sum_{j=1}^{p}\beta_j^2$ is the penalty function (that is, the L2 regularization term). The parameter λ (> 0) is the ridge parameter, and different values of λ correspond to different regression coefficients. When λ is 0, ridge regression is equivalent to ordinary least squares linear regression, and as λ approaches infinity, all regression coefficients are towards 0.

The general form solution of ridge regression model is as follows:

$$\widehat{\beta}^{ridge} = (X^TX + \lambda I)^{-1}X^Ty \quad （6）$$

In formula (6), X is an n×p matrix, y is a n-dimensional vector, I is a p×p unit matrix, and λ is the ridge parameter. By adding a positove constant identity matrix λI (λ > 0), formula (6) avoids the problem that X^TX is not of full rank and the singular matrix is not invertible due to the column linear correlation of X. Using the p×p unit matrix to add a “ridge” ensures that its matrix is invertible and the above equation has a solution. This equation is more stable in numerical calculation, avoids variable correlation, and makes the regression coefficient more accurate and stable.

3.3 Methods for sample collection and measuring of reconstituted tobacco

3.3.1 Sample collection

Due to the limitations of the continuous production process, the online measuring system cannot achieve real-time sampling. For this reason, the following steps are designed for sample data collection: 1) Before the sheet sample gose into the microwave sensor, manually divide the sample into areas and mark it with a marker pen in the designated area; 2) When the marked area reaches the position of the microwave sensor, the operator presses the “sampling” button to start the microwave data acquisition process; 3). After the microwave measurement is completed, manually remove the marked sheet part to prevent it from entering the cutter. Subsequently, cut off the sample of the marked part and number it, and the remaining sheet is sent to the cutter and processed according to the normal production process; 4) Send the sample sheet to the laboratory and measure the glycerol content by gas chromatography; 5) Correspond the microwave data and the gas chromatography results one by one to explore the glycerol content determination model.

3.3.2 Determination of glycerol content

Given that the microwave method is an indirect measurement technique, it is unable to directly and accurately determine the glycerol content in the absence of a glycerol content determination model. At present, chromatography is the industry-recognized standard method for measuring glycerol content in reconstituted tobacco. This study constructs a determination model for glycerol content by combining the measured values of glycerol in reconstituted tobacco obtained through gas chromatography with the corresponding microwave measurement values. Additionally, to evaluate the accuracy and reliability of the determination system proposed in this study, independent samples are used to validate the model.

4. Results and analysis

4.1 Establishment and evaluation of glycerol content determination model for thick paste method sheets

4.1.1 Variable definition description

On the basis of deeply discussion on the design principle and theoretical analysis of the dual-frequency microwave sensor, this study found that the variation of glycerol content in the reconstituted tobacco will affect key parameters such as resonant frequency and bandwidth. At the same time, microwave sensors with different frequencies may exhibit a certain degree of weak correlation in the measuring process. In view of this, this research needs to integrate the above parameters and their related variables systematically, and proposes new variables for regression analysis based on this foundation. Table 1 shows the variables closely associated with changes in the glycerol content of sheets.

Table 1 Variables definition description

Variables	Variable Definition	Minimum	Maximum	Median	Mean	Standard deviation
Y	Glycerol content/%	21.11	8.74	12.59	14.34	3.94
X1	Frequency shift of 1GHz resonator/MHz	5659.02	3012.14	3747.60	3896.32	662.46
X2	Bandwidth variation of 1GHz resonator/MHz	3383.77	1261.56	1978.42	2047.35	545.86
X3	Microwave value of 1GHz resonator	0.59	0.34	0.50	0.48	0.08
X4	Frequency shift of	31887.90	16677.25	21247.00	22098.45	3907.54

	4GHz resonator/MHz					
X5	Bandwidth variation of 4GHz resonator/MHz	13217.90	5854.71	8722.75	8864.71	1941.72
X6	Microwave value of 4GHz resonator	0.48	0.28	0.39	0.38	0.07
X7	X1·X3 Interaction term	3047.40	1192.69	1781.27	1869.28	461.50
X8	X2·X3 Interaction term	1822.17	433.89	1047.29	1015.43	404.46
X9	X4·X6 Interaction term	12529.96	5605.15	8153.34	8389.48	1757.40
X10	X5·X6 Interaction term	5425.83	1749.26	3275.50	3479.73	1217.37
X11	X3·X6 Interaction term	0.28	0.10	0.21	0.19	0.06

4.1.2 Ridge regression modeling analysis

Utilizing SPSS v16 software, an automatic correlation analysis was conducted on the input variables, and an appropriate ridge parameter was selected for calculation. Table 2 shows the ridge regression analysis results for the collected data of 23 sheet samples. After excluding the variables X2, X5, X7 and X10, the correlation coefficient of the ridge regression model is 0.982, the mean error is 0.33%, the standard deviation is 0.41%, indicating a high determination precision of the model.

Table 2 Ridge regression analysis results

Parameter	Non-standardized coefficient		Standard coefficient	t	Significance.
	B	Standard error			
Constant	59.213	16.290	NA	3.635	0.003
X1	0.027	0.003	6.827	10.449	0.000
X3	-41.181	19.158	-1.328	-2.150	0.051
X4	-0.005	0.001	-8.082	-5.956	0.000
X6	-302.842	86.310	-7.939	-3.509	0.004
X8	-0.033	0.007	-5.095	-4.640	0.000
X9	0.008	0.002	5.487	3.406	0.005
X11	366.527	60.406	8.946	6.068	0.000

Formula (7) is the corresponding ridge regression equation.

$$\hat{Y} = 59.213 + 0.027X_1 - 41.181X_3 - 0.005X_4 - 302.842X_6 - 0.033X_8 + 0.008X_9 + 366.527X_{11} \quad (7)$$

4.2 System determination repeatability and stability analysis

To ensure the accuracy of the determination results, the repeatability and stability of the system are of vital importance. In this study, gas chromatography was selected as the reference and verification method for the modeling of microwave system for glycerol in heat-not-burn reconstituted tobacco. Given that the microwave determination system designed in this paper is an online system, and repeatability tests require multiple measurements of the same sample, which is difficult to achieve in the online detection process. Therefore, in this paper, the offline sampling method was used to perform 8 repeated measurements on each group of 5 independent samples to evaluate the repeatability of the detection system. As shown in Table 3, the repeatability test results show that the repeated measurement standard deviation of the 5 groups of samples is less than 0.1%, and the coefficient of variation is less than 1%, which fully demonstrates that the determination system has high measurement precision.

Table 3 Results of repeatability test

No.	Content of glycerol by microwave method /%				
	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5
1	12.65	12.33	11.85	11.98	11.17
2	12.51	12.23	11.68	12.03	11.23
3	12.49	12.34	11.74	11.88	11.20
4	12.53	12.36	11.71	11.97	11.25
5	12.58	12.29	11.76	12.00	11.31
6	12.62	12.31	11.81	12.08	11.35
7	12.56	12.32	11.83	11.92	11.15
8	12.68	12.28	11.75	11.99	11.24
mean	12.58	12.31	11.77	11.98	11.24
Standard deviation	0.07	0.04	0.06	0.06	0.07
Coefficient of variation	0.54	0.33	0.50	0.52	0.60

The stability of the online determination system is crucial for maintaining consistent performance during the long-term operation. In the production processing of heat-not-burn reconstituted tobacco, due to steps such as high-temperature drying, the environment of the production is usually high-temperature and high-humidity. Such conditions may cause drift in the microwave online system. To address this issue, the following strategy was adopted stability testing in this paper: install the microwave online system in the production workshop and conduct testing in offline sampling mode. Ten groups of sheet samples with similar glycerol content were selected, with one sample measured every 1 h, and the sample was determinated with gas chromatography in laboratory. Figure 4 shows the offset results of the system. In the figure, rectangular marks represent the hourly data measurement points of the microwave system, while triangular marks represent the measurement results of the same sample measured by gas chromatography. It can be clearly observed from the figure that during the long-term operation of the system, the deviation between the glycerol content measured by the microwave method and the result of gas chromatography is minor, with the maximum deviation of only 0.64%, which meets the determination accuracy requirements of the model. Moreover, no drift phenomenon of measurement error over time is observed. The test result validate the stability of the microwave system for glycerol content measuring in heat-not-burn reconstituted tobacco during long-term measurement and ensure that the accuracy of the measurement results is not affected by time.

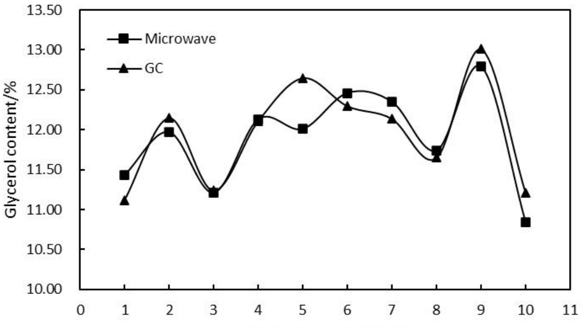


Fig. 4 Offset results of the microwave online system over time

4.3 Online determination and analysis of glycerol in heat-not-burn reconstituted tobacco

Based on the above established glycerol determination model, as described in section 2.4, 20 groups of samples were randomly marked and selected from the production of a certain brand. The glycerol contents were determined by microwave system and gas chromatography respectively. Table 4 shows the measured results. According to the analysis of the data in the table, the mean error between the microwave method and the Gas Chromatography method is 0.08%. To further verify the accuracy, a paired t-test was carried out between the two results using SPSS V16 software. At a 95% confidence level, the mean difference is 0.04313(confidence interval [-0.26402,0.09702]), and the p-value is 0.345(> 0.05), indicating no statistically significant difference in the measurement results of glycerol content between the two methods.

Table 4 Determination of glycerol content with different methods

No.	Microwave measurement /%	GC measurement /%	Error/%	No.	Microwave measurement /%	GC measurement /%	Error/%
1	10.57	10.09	-0.48	11	10.58	10.31	-0.27
2	11.01	11.23	0.22	12	11.34	11.01	-0.33
3	10.89	11.12	0.23	13	12.34	11.99	-0.35
4	11.59	11.41	-0.18	14	12.64	12.87	0.23
5	11.97	12.31	0.34	15	13.14	13.45	0.31
6	10.07	10.74	0.67	16	13.78	13.65	-0.13
7	12.35	11.85	-0.50	17	12.78	12.96	0.18
8	11.76	12.34	0.58	18	10.28	10.65	0.37
9	12.14	12.37	0.23	19	11.46	12.25	0.79
10	13.24	12.89	-0.35	20	12.34	12.45	0.11

In conclusion, the online determination system of the atomizing agent content in the heat-not-burn reconstituted tobacco, designed and developed in this study based on microwave technology, not only exhibits higher detection accuracy, but also shows no significant difference when compared to existing conventional methods. The system can fully capable of meeting the needs of online sheet production and provides solid technical support for the enterprise's large-scale production.

5. Conclusions

In this paper, a dual-band microwave sensor is designed and developed based on the microwave technology. On this basis, an online device of atomizer content in heat-not-burn reconstituted tobacco is developed. Taking gas chromatography as a benchmark method for modeling and validation, this study used ridge regression analysis to establish a determination model for glycerol content. The correlation coefficient of the model is as high as 0.982, and the mean error is only 0.33% , and the standard deviation is 0.41% . In addition, the results of repeatability and stability test shows that the system has high precision and stability, and does not experience drift of measurement errors due to time, It can fully meet the needs of online production, and provide strong technical support for quality assurance and determination efficiency improvement of heat-not-burn reconstituted tobacco.

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