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Microwave Drying of Anthracite: A Parameter Optimized by Response Surface Methodology

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Abstract Response surface methodology, based on Central composite design, was applied to obtain the optimized parameters for microwave drying on anthracite. The microwave power level, drying time, and sample mass were selected as the independent variables for the experiments, while the dehydration ratio was selected as the response affected by the independent variables. The significance of these three variables are in reverse order: drying time ≫ sample mass > power level. The interactions between drying time and power level, power level, and sample mass were significant; the interaction between drying time and sample mass was insignificant. The optimization conditions were the following: power level of 682.07 W; drying time of 2.98 min; and sample mass of 49.19 g. The maximal effectiveness ratio is 1.452 kg/kW h under the optimized condition. The verification experiment indicated that the experimental results were in good agreement with the predicted values, which has only a +0.57% deviation.

Keywords Anthracite · Microwave drying · Dehydration ratio · Optimization · Response surface methodology (RSM)

الخلاصة

في الحصول على (CCD) المعتمدة على تصميم التركيب المركزي (RSM)لقد تم تطبيق منهجية استجابة السطح العوامل المحسنة في تجفيف الفحم الصلب باستخدام المايكروويف. وتم اختبار مستوى قوة المايكروويف وزمن التجفيف و وزن العينة كمتغيرات مستقلة للتجربة، بينما اختيرت نسبة التجفيف على أنها الاستجابة المتأثرة بالمتغيرات المستقلة. إن أهمية هذه المتغيرات الثلاثة عكسيا هي: زمن التجفيف>> وزن العينة > مستوى الطاقة. كان التداخل بين زمن التجفيف و مستوى الطاقة و بين مستوى الطاقة و وزن العينة هاما في حين كان التداخل بين زمن التجفيف و وزن العينة غير هام. إن الظروف المحسنة كانت كالتالي: 682.06 واط مستوى طاقة, 2.98 دقيقة زمن تجفيف و 49.19 غم وزن عينة. لقد كانت نسبة التأثير القصوى هي 1.452 كغم/كيلوواط في الظروف المحسنة. إن تثبيت التجربة دل على أن النتائج التجريبية كانت بتوافق جيد مع القيم المخمنة و بنسبة حيود 0.57% فقط.

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1 Introduction

Anthracite is the highest coalification coal, whose fixed dry basis carbon content is about 90%. Due to its high combustion heating value, anthracite is commonly used as power fuel [1], sintering and casting fuel [2], reducing agent in iron ore blast furnace smelting [3,4], and raw material of coking, gasification, and liquefaction [5,6]. In anthracite, moisture is the major component of volatile matter. When used as a fuel or industrial raw material, in order to reduce the thermal losses (transported and exchanged by vapor) and increase the latent heat of materials, anthracite need to be dried to remove most moisture. In addition, high moisture content can have a great tendency to combust spontaneously. Thus, the removal of moisture from anthracite is an important pre-processing. Currently, solar drying and convective drying are the widely used technique for coal dehydration. Solar drying requires a big yard and yields a low drying rate. Convective drying ordinarily uses the waste flue gas as a thermal medium. When processing, the vapor diffusion direction inside the material is exactly opposite to the temperature gradient, which can lower the heat transport efficiency and slow down the vapor diffusion speed [7,8]. Thus the convective drying also gets a poor drying rate. Therefore, the development of a highly efficient anthracite drying method has been the subject of intensive research.

Microwave is a kind of electromagnetic wave, with frequency between 3×10^8 and 3×10^{11} Hz. The outstanding features of microwave heating are rapidity and selectivity. The water-specific inductive capacity of 60–78 is much greater than common ores. When irradiated in a microwave field, moisture always quickly absorbs electromagnetic wave and is rapidly transferred to vapor. In addition, the other distinct advantages of microwave drying include no need of accessory equipment and almost no pollution release. Many literatures referred to the microwave drying of woods [9], vegetables and fruits [10,11], and ore [12].

Response surface methodology (RSM) is an optimization method of asymptotic analysis. The RSM was developed in the 1930s and formally proposed by G.E.P. Box in the 1950s [13]. At first RSM was generally used in chemical industry in 1950s, whose purpose is to determine the optimal operation and obtain an optimal or expected response [14]. Several previous researchers have proved that RSM is a powerful statistical tool in designing experiments due to its simplicity [15], fewer experiments required, and the consideration of the interactions between variables [16]. There is no literature referring to parameter optimization of microwave drying of anthracite. In this study, the ultimate aim was to investigate the effects of microwave power level, drying time, and sample mass on the dehydration ratio of anthracite. RSM was employed to optimize the drying conditions.

2 Experimental

2.1 Anthracite

Anthracite was obtained from Yunnan Coal Chemical Industry Group Co., Ltd, Yunnan Province, China. The proximate analysis of anthracite is shown in Table 1, giving 24.25% moisture and volatile content, 12.54% ash content, and 63.21% fixed carbon. The initial moisture content was measured using oven-drying method [17], see Table 2. The results show the initial moisture content is from 0.208 to 0.210 kg/kg w.b., and the average value is 0.209 kg/kg w.b.

Table 1 Proximate analysis of anthracite

Component	Ash	Moisture and volatile matter	Fixed carbon
Mass percent (wt%)	12.54	24.25	63.21

Table 2 Initial moisture content of anthracite

No.	Before dry (g)	After dry (g)	Moisture content (kg/kg w.b.)
1	5.000	3.956	0.209
2	5.000	3.949	0.210
3	5.000	3.960	0.208
Average			0.209

2.2 Drying Apparatus and Procedure

The drying experiments were conducted in a refined domestic microwave oven (Glanz G80D23ESL, China) with technical features of 230 V, 50 Hz and frequency of 2,450 MHz (a wavelength of 12.24 cm), See Fig. 1. The microwave oven was the standard one with the capability of operating at continuous power levels from 0 to 800 W. The microwave power level and processing time were adjusted with the aid of a manual control facility located on the microwave oven. The mass loss was measured by a thermo-gravimetric balance, which had a precision of 0.001 g, and was connected to the computer. A software (Shanghai Precision Scientific Instrument Company) was used for data collection. The hang stage material was made of PTFE, a low dielectric material. When caring out the microwave drying experiment, a specific amount of anthracite was put onto the hang stage, and then the microwave power was set to the desired level, and the experiment started. Prior to this procedure, a temperature rising experiment was conducted to determine the microwave absorbing property of anthracite, see Fig. 2. It shows that 50 g sample was heated to 110°C in 90 s under 700 W power level, illustrating a relative good microwave absorbing property of anthracite.

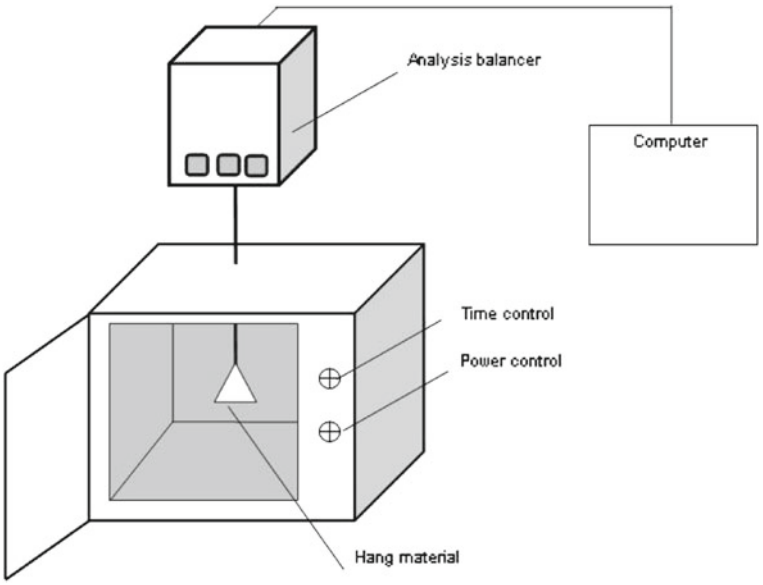

Fig. 1 Connection diagram of microwave drying equipment

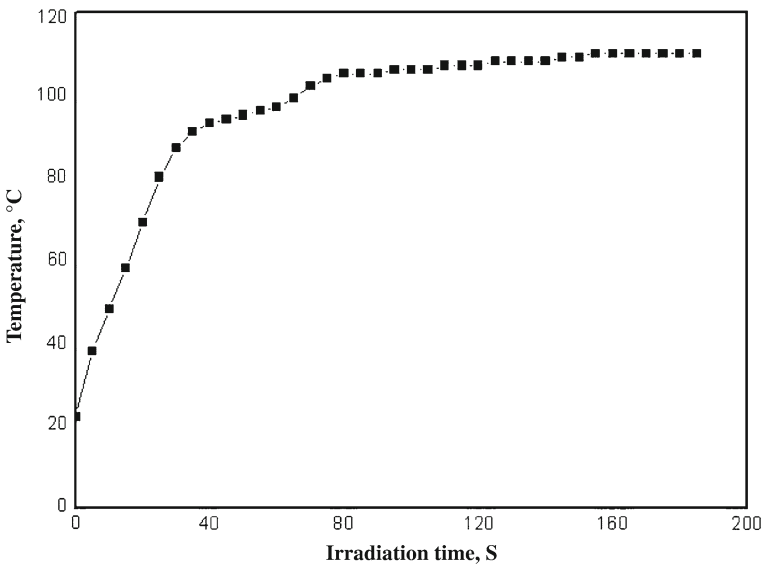

Fig. 2 Temperature raising curve of anthracite under the microwave power of 700 W

Table 3 Independent variables and their levels used for CCD

Independent variables	Symbol	Coded variable levels				
		−1.682	−1	0	+1	+1.682
Power level (W)	x_1	324.55	420	560	700	795.45
Drying time (min)	x_2	0.32	1	2	3	3.68
Sample mass (g)	x_3	9.77	20	35	50	60.23

2.3 Dehydration Ratio Analysis

The dehydration ration of anthracite was calculated using the Eq. (1).

$$\text{Dehydration ratio} = \frac{M_0 - M_i}{M_0}, \tag{1}$$

where M_0 is the initial moisture content and M_i is the moisture content at specific time.

2.4 Experiments Design

Central composite design (CCD) was applied to investigate the effects of three independent variables, i.e. microwave power level χ_1 (W), drying time χ_2 (min), and sample massχ_3 (g), on dehydration ratio (See Table 3). The chosen independent variables used in this study were coded according to Eq. (2).

$$\chi_i = \frac{\chi_i - \chi_0}{\Delta\chi}, \tag{2}$$

where χ_i is the dimensionless value of the ith independent variable, χ_0 is the value of χ_i at the center point, and $\Delta\chi$ is the step change value. The dehydration ratio of anthracite was taken as the response in the designed experiments. A total of 20 experiments consisting of 8 factorial points, 6 axial points, and 6 replicates at the central points were performed. Experimental data obtained from the CCD model experiments can be described in Eq. (3)

$$Y = \beta_0 + \sum_{i=1}^{n} \beta_i \chi_i + \sum_{i=1}^{n} \beta_{ii} \chi_i^2 + \sum_{i<j} \beta_{ij} \chi_i \chi_j \tag{3}$$

where β_0 is a constant; and β_i, β_{ii}, β_{ij} are the linear coefficient, quadratic coefficient, and interactive coefficients, respectively. The analyses of variance (ANOVA) and response surface drawing were performed using the Design Expert Software (Version 7.15) from Stat-Ease inc., USA. The optimized experimental parameters were obtained by using the software's numerical and graphical optimization functions.

3 Results and Discussion

3.1 Response Analysis and Interpretation

Table 4 shows the results of the experiments conducted. The results of dehydration ratio were found ranging from 3.03 to 66.72%. When the dehydration ratio has the lowest value of 3.03% (Run 11), the experimental conditions are power level of 560 W, drying time of 0.32 min, and sample mass of 35 g. The independent variables of x_1 and x_3 were at the 0 level, and the drying time x_2 was at the −1.682 level, indicating the irradiation time might has a critical effect on microwave drying of anthracite. Correspondingly, when the dehydration ratio gets a highest value of 66.72%, drying time x_2 was at the +1.682 level. Run 15 to 20 belong to the central point experiments which were used to determine the experimental error [18]. According to the sequential model, the sum of squares can be obtained. The models were selected based on the highest order polynomials where the additional terms are significant and the model is not aliased [19]. The quadratic model was selected for dehydration ratio determination as suggested by the software.

Table 4 Experiment results from CCD

Experiment no.	Variables			Response value
	Power level χ_1 (W)	Drying time χ_2 (min)	Sample mass χ_3 (g)	Dehydration ratio Y (%)
1	420(−1)	1(−1)	20(−1)	22.96
2	700(+1)	1(−1)	20(−1)	17.65
3	420(−1)	3(+1)	20(−1)	48.82
4	700(+1)	3(+1)	20(−1)	63.38
5	420(−1)	1(−1)	50(+1)	6.59
6	700(+1)	1(−1)	50(+1)	12.59
7	420(−1)	3(+1)	50(+1)	29.65
8	700(+1)	3(+1)	50(+1)	59.29
9	324.55(−1.682)	2(0)	35(0)	12.77
10	795.45(+1.682)	2(0)	35(0)	37.80
11	560(0)	0.32(−1.682)	35(0)	3.03
12	560(0)	3.68(+1.682)	35(0)	66.72
13	560(0)	2(0)	9.77(−1.682)	62.08
14	560(0)	2(0)	60.23(+1.682)	35.45
15	560(0)	2(0)	35(0)	24.20
16	560(0)	2(0)	35(0)	24.03
17	560(0)	2(0)	35(0)	24.71
18	560(0)	2(0)	35(0)	24.20
19	560(0)	2(0)	35(0)	24.54
20	560(0)	2(0)	35(0)	24.54

Table 5 Analysis of valance (ANOVA) for response surface quadratic model

Source	Sum of squares	Degree of freedom	Mean square	F value	Prob > F
Model	6932.3930	9	770.2659	77.9385	<0.0001
χ_1	554.0396	1	554.0396	56.0599	<0.0001
χ_2	4520.3710	1	4520.3710	457.3888	<0.0001
χ_3	586.2243	1	586.2243	59.3165	<0.0001
$\chi_1\chi_2$	236.6400	1	236.6400	23.9442	0.0006
$\chi_1\chi_3$	87.0540	1	87.0540	8.8085	0.0141
$\chi_2\chi_3$	0.4186	1	0.4186	0.0424	0.8411
χ_1^2	5.3350	1	5.3350	0.5398	0.4794
χ_2^2	111.5478	1	111.5478	11.2868	0.0072
χ_3^2	852.8919	1	852.8919	86.2989	<0.0001
Residual	98.8300	10	9.8830	–	–

The final empirical model in terms of coded factors after including the insignificant terms is shown in Eq. (4).

$$Y = 24.51 + 6.37x_1 + 18.19x_2 - 6.55x_3 + 5.44x_1x_2 + 3.30x_1x_3 - 0.23x_2x_3 - 0.61x_1^2 + 2.78x_2^2 + 7.69x_3^2 \quad (4)$$

The quality of the model developed was evaluated based on the correlation coefficiency value. Values of "Prob > F" less than 0.0500 indicate model terms are significant. In this case, χ_1, χ_2, χ_3, and the interaction terms $\chi_1\chi_2$, $\chi_1\chi_3$, χ_2^2, and χ_3^2 were significant model terms of the response. Values of "Prob > F" greater than 0.1000 indicate that the model terms are not significant. The Values of "Prob > F" of x_2x_3 and x_1^2 are 0.8411 and 0.4794, respectively.

The closer the R^2 value to unity and the smaller the standard deviation, the more accurate the response could be predicted by the model. The R^2 value for Eq. (4) was found to be 0.986, indicating that there was a good agreement between the experimental data and the predicted moisture content from models.

The results obtained from the analysis of variance (ANOVA) proved the validity of the model. The ANOVA for the quadratic model in determining moisture content is listed in Table 5. The model F value of 77.94 implied that the model was significant. There is only a 0.01% chance that a "Model F-Value" this large could occur due to noise. The "Pred R-Squared" of 0.894 is in reasonable agreement with the "Adj R-Squared" of 0.973, implying that the above models were adequate to predict the moisture content within the reasonable range of the variables studied. The check of model adequacy is an important part of the data analysis procedure. "Adeq Precision" measures the signal to noise ratio. For a fixed model, adequate precision measurement of the signal to noise ratio is necessary and a ratio greater than 4 is desirable. In the quadratic model, the ratio of 29.725 indicates adequate signal for the model to be used to navigate the design space.

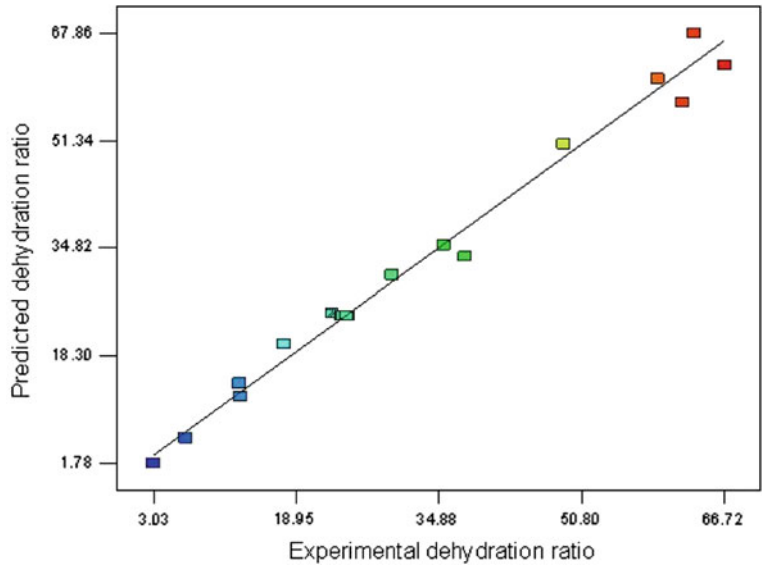

Fig. 3 Predicted versus experimental dehydration ratio

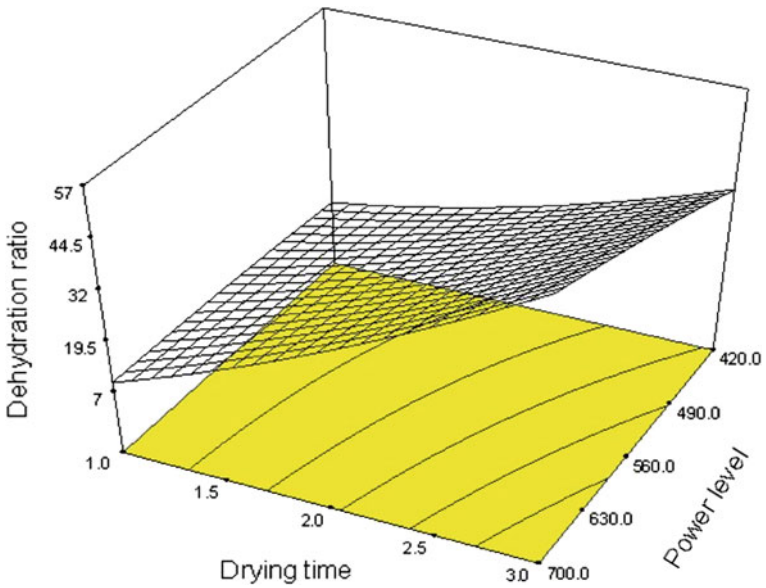

Fig. 4 Response surface and contour plot of power level versus drying time (sample mass was fixed at zero level)

Figure 3 shows the predicated dehydration ratio versus the experimental values. Actual values were derived from the measured response data for a particular run, and the predicted values were evaluated from the model and generated by using the approximating functions. As can be seen, the predicated value obtained was close to the experimental values, indicating that the model developed was successful in capturing the interaction between the variables and the moisture content. Under the designed conditions, a few experiment results show little difference with the predict values.

The three-dimensional response surfaces, which were constructed to show the effects of the three variables on Y, were shown in Figs. 4, 5, and 6. As can be seen from Figs. 4, 5, and 6 the drying time is the most significant variable affecting the dehydration ratio, while there were minor significant contribution of power level and sample mass. The results agree with the ANOVA (Table 5), and the F value of drying time, power level, and sample mass are 457.3888, 56.0599, and 59.3165, respectively. Joint the analysis of ANOVA and contour plots, the interaction between drying time and power level is more distinct than that of between power

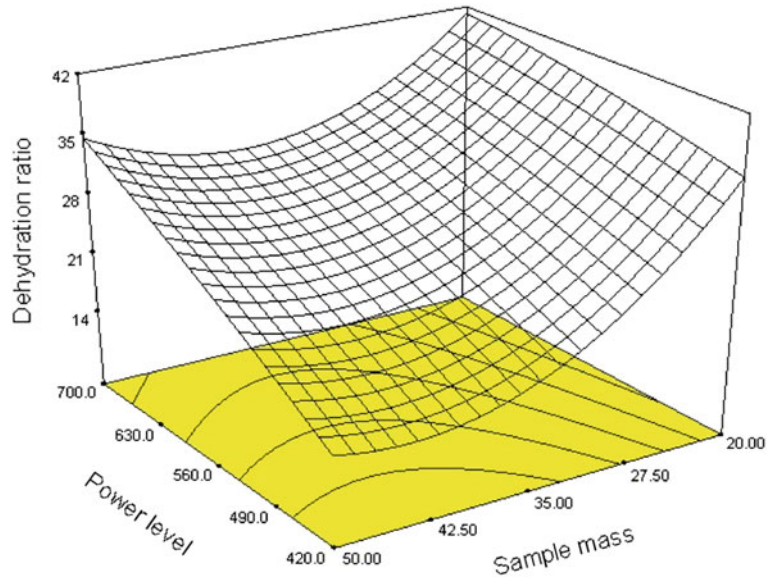

Fig. 5 Response surface and contour plot of power level versus sample mass (drying time was fixed at zero level)

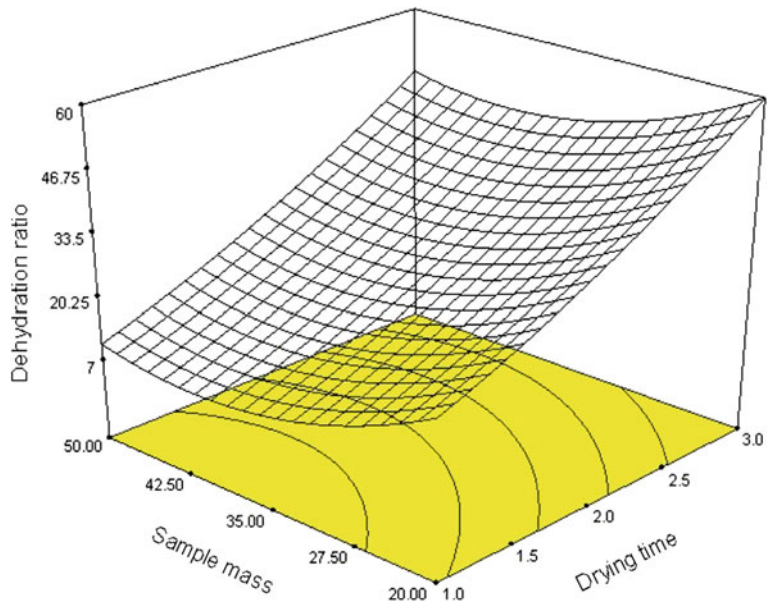

Fig. 6 Response surface and contour plot of drying time versus sample mass (power level was fixed at zero level)

level and sample mass. The interaction between drying time and sample mass was almost absent, and the F value is 0.0424. The finding is similar to results in the literature [15,16].

Figure 7 shows different factors affecting the drying process in term of dehydration ratio. The cubic graphs shows again that the drying time has greatest effect on dehydration ratio, and the significance of these three variables are in reverse order: drying time $\gg$ sample mass $>$ power level. The maximum dehydration ratio 67.86% was located at the vertex, indicating another index should be included to optimize the process parameters.

3.2 Parameter Optimization

The final moisture content of anthracite should be less than 0.1 kg/kg w.b. (Coal Chemical Industry Group Co., Ltd). So, the dehydration ratio in this work has to be larger than 57.9%. According to the analysis above, drying

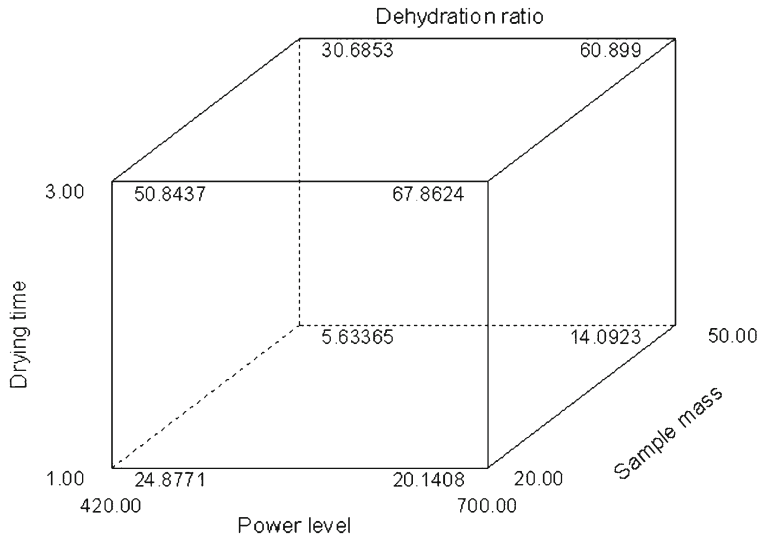

Fig. 7 Cubic graphs for different factors affecting the drying process in term of dehydration ratio

Table 6 Single condition optimized results

No.	Power level (W)	Drying time (min)	Sample mass (g)	η (kg/kW h)	Credibility	Predicted value
1	533.65 (Min)	3.00	20.17	0.756	1.000	58.04
2	698.98	2.69 (Min)	20.32	0.648	1.000	58.49
3	700.00	3.00	50 (Max)	1.429	1.000	60.90
4	688.00	2.93	20.75	0.618	1.000	64.26 (Max)

Table 7 Final optimized results: effectiveness ratio, predicted and experimental dehydration ratio under the optimized condition

Independent variables			η (kg/kW h)	Credibility	Dehydration ratio (%)		
Power level (W)	Drying time (min)	Sample mass (g)			Predicted value	Experimental value	Deviation
682.07	2.98	49.19	1.452 (Max)	1.000	57.90	58.23	+0.57%

time is the most significant variable, so the optimization range of drying time is narrow. An effectiveness ratio η was used to evaluate the economical efficiency of optimal conditions, see Eq. (5).

$$\eta = \frac{\text{Mass}}{\text{Power} \times \text{Time}}, \tag{5}$$

where η is effectiveness ratio, in unit of kg water/kW h; mass, power, and time were the optimized variables given by software. Under the condition of dehydration, the ratio is larger than 57.9%; the optimization of each variable and the optimized results are given in Table 6. As can be seen from Table 6, the optimized independent variables are the following: the minimum power level is 533.65 W, the minimum drying time is 2.69 min, and the maximum sample mass is 50 g. Under an appropriate condition, the dehydration ratio can reach the maximum value 64.26%. The final optimized results are shown in Table 7. Under those conditions, the maximum η of 1.452 and a dehydration ratio of 57.90% were obtained. Putting the above parameters into experiment, the actual dehydrate value of 58.23% was obtained, compared with the predicted value of 57.90%, giving us the deviation of +0.57%, indicating that the process under the conditions of the experimental data approximates to the predictive value.

4 Conclusion

The microwave drying of anthracite was studied in this work. A parameter optimization by RSM was investigated. The final empirical model including significant and insignificant terms was shown in

Eq. (4). Through analyzing the quadratic drying model, the significance of three variables and the interaction between each other were given by software. The drying time yielded the greatest impact on dehydration ratio, and the significance of these three variables are in reverse order: drying time $\gg$ sample mass > power level. The interactions between drying time and power level, power level and sample mass were significant; the interaction between drying time and sample mass was insignificant.

The optimized parameters of microwave dehydration are shown in the following: microwave power is of 682.07 W, radiation duration is of 2.98 min, amount of material is of 49.19 g. Then, the most efficient treatment of 1.452 kg/kW h was achieved, giving the deviation +0.57%.

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