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Optimization of microwave heating thickness for spent automobile catalyst

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Abstract: A new method was developed to optimize the microwave heating thickness of the spent automobile catalyst in order to improve the uniform distribution of the temperature field. The average penetration depth and the microwave heating thickness of the spent automobile catalyst were calculated by Gauss model and numerical calculation based on dielectric loss tangent and reflection loss. The results showed that the spent automobile catalyst was a medium loss material. The average penetration depth was 1.11 m from room temperature to 800 °C. The optimum microwave heating thickness of the spent automobile catalyst was about 0.83 m or 0.75 times of the average penetration depth. Industrial application analysis indicated that the optimization of heating thickness could improve the uniform distribution of the temperature field and reduce energy consumption.

Key words: spent automobile catalyst; microwave; heating thickness; optimization

1 Introduction

Automobile catalyst is mainly used to convert CO, CH_x and NO_x into non-toxic CO_2, H_2O and N_2, and it consists of carrier and catalytic active substances. The active substances are mainly platinum group metals (PGM), such as Pt, Pd, and Rh [1,2]. More than 60% of PGM are used in automobile exhaust industry. With the abandonment of a large number of automobiles, lots of the spent automobile catalysts accumulated in many parts of the suburbs. This leads to environmental pollution and waste of scare resources [3]. Therefore, the recovery of PGM from the spent automobile catalyst has attracted a lot of attention.

The recovery methods of PGM from the spent automobile catalyst include pyrometallurgical and hydrometallurgical processes. Especially, microwave technology has attracted much attention in the field of recovering precious metals from the spent automobile catalyst in recent years [4−6], because it can improve the leaching efficiency, reduce the roasting temperature and shorten the leaching time [7,8]. However, one of the major problems in microwave heating process is uneven distribution of

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temperature field [9,10], which causes hot spots and cold centers. The hot spots and cold centers would result in overheating or incomplete reaction, high energy consumption, low energy efficiency, and so on [11]. The uneven distribution of temperature field is caused by non-uniform electromagnetic field inside microwave cavity and uneven absorption of microwave energy by the spent automobile catalyst [12]. Therefore, it is the research focus to realize the uniform distribution of temperature field in microwave heating.

The optimization of the microwave heating thickness is an effective method to solve the uneven temperature distribution [13], because too small thickness would cause hot spots and high energy-consumption, and too large thickness would cause cold centers [14]. In literatures, experiments and simulations were wildly used to optimize microwave heating thickness [15−18]. However, the processes are tedious and time-consuming. SHANG et al [19] obtained the multiple values of the optimized thickness for silica. However, they didn't suitable for the actual production process. Therefore, it is necessary to develop an effective method to optimize the microwave heating thickness based on the material nature.

This work was to develop an optimization method of microwave heating thickness. The dielectric properties of the spent automobile catalyst were measured from room temperature to 800 °C at 2450 MHz. The microwave absorption characteristics were studied by theoretical analysis and heating experiment. Based on the absorption characteristics and dielectric properties, the optimum microwave heating thickness of the spent automobile catalyst was obtained by numerical calculation from room temperature to 800 °C at 2450 MHz.

2 Experimental

2.1 Material

The spent automobile catalyst was crushed and milled into powder using a ball mill. The volume cumulative distribution and volume frequency distribution were examined using a JL−1177A laser particle analyzer. As shown in Fig. 1, the area average particle size and volume average particle size of the powder sample are 3.57 μm and 25.63 μm, respectively.

X-ray diffraction (XRD) pattern of the spent automobile catalyst is shown in Fig. 2. The main components of the spent automobile catalyst were cordierite, γ-Al_2O_3 and CeO. The chemical composition of the spent automobile catalyst was confirmed by X-ray fluorescence (Axios PW4400/40, Panalytical, Netherlands). The normalized element contents of platinum and rhodium are 0.11 and 0.04 wt.%, respectively (Table 1).

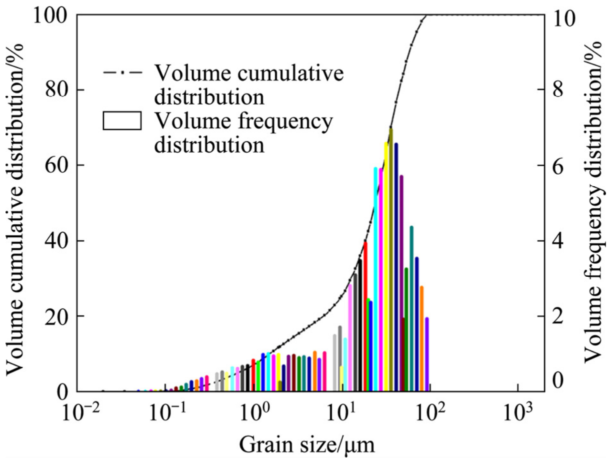

Fig. 1 Volume cumulative distribution and volume frequency distribution of spent automobile catalyst

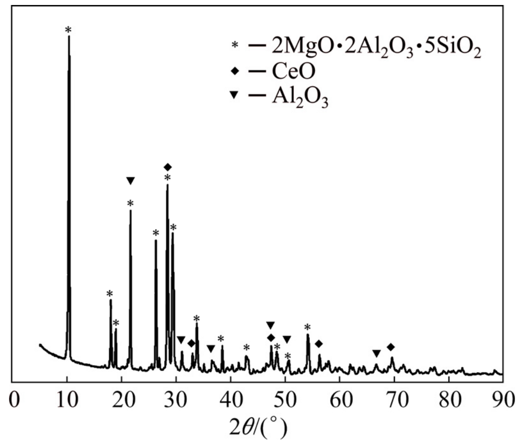

Fig. 2 XRD pattern of spent automobile catalyst

Table 1 Chemical composition of spent automobile catalyst (wt.%)

O	Al	Si	Mg	Ce	Fe	Ca	Pt	Rh
48.06	25.89	15.01	5.11	3.88	0.53	0.33	0.11	0.04

2.2 Measurement of dielectric properties

Dielectric properties are the principal parameters that determine how the energy is absorbed and transmitted inside a material. The dielectric properties can be expressed in the followed form [20]: $\varepsilon_r=\varepsilon_r'-j\varepsilon_r''$, where relative dielectric constant (ε_r') represents the storing ability of microwave energy, and relative dielectric loss factor (ε_r'') represents the dissipating ability of microwave energy inside a material.

The dielectric properties were measured by the cavity perturbation method [21,22]. The measurement system includes induction heating equipment, circulating water cooling equipment, pneumatic equipment, TM_{0n0} cylindrical cavity, vector network analyzer (Keysight Technologies, Agilent N5230C), computer and test software. The test software is used to calculate and calibrate the date collected from the cavity perturbation. The powder of spent automobile catalyst was dried at 100 °C for 10 h in a drying oven and was put into a quartz tube with length of 500 mm and inside diameter of 28 mm. The measurements were conducted every 50 °C in the range from room temperature to 800 °C. The dielectric properties were measured three times, and the average data were used. The entire measurement process was in an air atmosphere. The mass of sample was weighed by analytical balance. The volume of sample was calculated based on the values of radius and height. The density of spent automobile catalyst could be calculated (1.48 g/cm^3). The mass didn't change before and after the measurement of dielectric properties.

2.3 Microwave heating equipment

The microwave heating equipment in the present work is designed and fabricated in the Key Laboratory of Unconventional Metallurgy of the Ministry of Education, Kunming University of Science and Technology (Fig. 3). The inner dimensions of the microwave cavity are: 420 mm in length, 260 mm in width and 260 mm in height. The thermal insulation material is placed in the microwave cavity to reduce heat loss. The power of the microwave reactor is adjustable from 0 to 4.8 kW at 2.45 GHz. The temperature was measured by a K-type thermocouple connected to the control system and inserted from the top of microwave cavity. The thermocouple is 1000 mm in length and 3 mm in diameter, with the temperature range of 0−1250 °C and the measurement precision of ±0.5 °C. The sample is placed in the alumina crucible, and the thermocouple was inserted into the sample during the heating process.

Fig. 3 Photo of microwave heating equipment

2.4 Reflection loss and power penetration depth

Reflection loss is a significant parameter to evaluate microwave absorbing characteristics. According to transmission line theory, microwave irradiates vertically to absorber slab backed by an ideal conductor. The reflection loss (L_R) can be summarized by the following equations [23,24]:

$$Z_{in}=Z_0\sqrt{\frac{\mu_r}{\varepsilon_r}}\tanh\left[j\left(\frac{2\pi fd}{c}\right)\sqrt{\mu_r\varepsilon_r}\right] \tag{1}$$

$$L_R=20\lg\left|\frac{Z_{in}-Z_0}{Z_{in}+Z_0}\right| \tag{2}$$

where Z_{in} is the normalized input impedance and Z_0 is the impedance of free apace; μ_r is relative complex permeability and is assumed to be 1 in this study; d is the thickness of absorber slab; c is the speed of light in free space; f is the frequency of microwave; L_R is a ratio of the reflection power to the incident power. A lower value of L_R stands for a better microwave absorbing characteristics.

The power penetration depth (D_p) is a very important factor in the design and scale-up of a microwave heating system. It is defined as the distance from the surface into material at which the incident wave power density attenuates to e^{-1} of its surface value, and can be calculated by [25,26]

$$D_p=\frac{c}{2\pi f\sqrt{2\varepsilon_r'\left[\sqrt{1+\left(\varepsilon_r''/\varepsilon_r'\right)^2}-1\right]}} \tag{3}$$

3 Results and discussion

3.1 Dielectric properties of spent automobile catalyst

The influence of temperature on the dielectric properties of the spent automobile catalyst at 2.45 GHz is shown in Fig. 4. The dielectric constant decreased from room temperature to 200 °C and followed by a slight fluctuation as the temperature increased to 800 °C in Fig. 4(a). The greater thermal agitation disturbed the alignment of molecular dipoles with the electromagnetic field as temperature increased and reduced the polarization in the range from room temperature to 200 °C. The value of the dielectric constant varied from 3.3995 at 200 °C to 3.5983 at 800 °C, and its arithmetic mean was 3.476, indicating that spent automobile catalyst was weakly polar. These results suggested that temperature had a little effect on the dielectric constant and polarity of the spent automobile catalyst, which was attributed to insensitivity of its main chemical components to temperature. As shown in Fig. 4(b), the dielectric loss factor decreased slightly from room temperature to 200 °C (its minimum value was 0.0176 at 200 °C), and then increased as temperature increased to 800 °C (its maximum value was 0.1166 at 800 °C). So, the temperature had a significant influence on the dielectric loss factor of the spent automobile catalyst at 2.45 GHz. The dielectric loss factor characterizes the ability of a material to dissipate electromagnetic energy into heat, which results from dipole loss and conduction loss [27,28]. Dipole is an important physical model in the study of dielectric materials. Generally, a polar molecule can be equivalent to a dipole. The dipole loss decreases with the increase of temperature, and the conduction loss increases with the increase of temperature. When one of the two loss mechanism is dominant, the trend of the dielectric loss factor corresponds to this mechanism. The curve of dielectric loss factor may show a valley feature when the contribution of the two loss mechanisms are equal [22]. Figure 4(b) indicated that the dipole loss was dominant from room temperature to 200 °C and the conduction loss was dominant from 250 to 800 °C [29]. Hence, the smaller drop of the dielectric loss factor was attributed to smaller decrease of the dielectric constant from room temperature to 200 °C. The increase of the dielectric loss factor from 250 to 800 °C might be caused by the PGM in spent automobile catalyst, because the outer electrons of PGM became unstable with the increase of temperature. As a consequence, the temperature-dependent electrical conductivity could enhance the temperature-dependent dielectric loss factor of spent automobile catalyst [22].

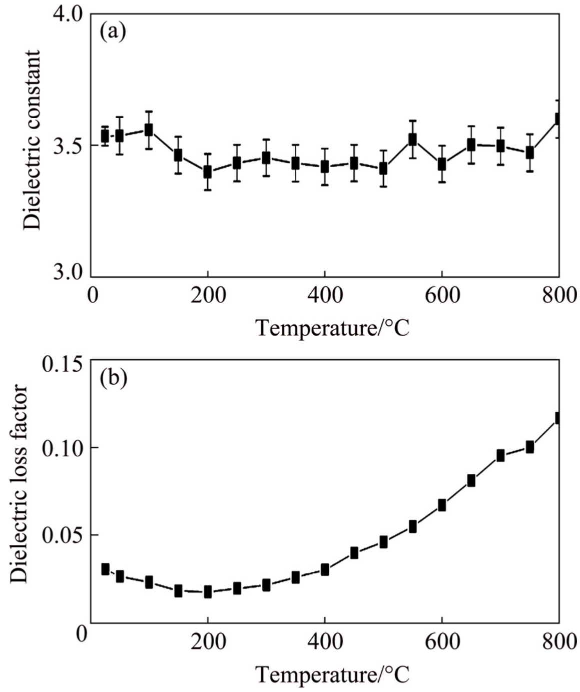

Fig. 4 Temperature dependence of dielectric properties at 2.45 GHz: (a) Dielectric constant; (b) Dielectric loss factor

3.2 Microwave absorbing characteristics of spent automobile catalyst

Dielectric loss tangent ($\tan\delta=\varepsilon_r''/\varepsilon_r'$) is an intuitive parameter to describe the ability of a material to convert microwave energy into heat [30]. In general, the microwave absorbing ability is greater when the dielectric loss tangent is higher. LAYBOURN et al [31] classifies materials by the dielectric loss tangent. The $\tan\delta$ of the low loss materials is smaller than 3×10^{-4}. The $\tan\delta$ of the medium loss materials is between 3×10^{-4} and 3×10^{-2}. The $\tan\delta$ of the high loss materials is larger than 3×10^{-2}. Figure 5 shows the temperature dependence of $\tan\delta$ at 2.45 GHz. The dielectric loss tangent of the spent automobile catalyst decreased from room temperature to 200 °C, and then increased with temperature to 800 °C. The minimum value of the $\tan\delta$ is 0.0052 at 200 °C while the maximum is 0.0324 at 800 °C. Hence, the spent automobile catalyst is medium loss material from room temperature to 750 °C and high loss material from 750 to 800 °C.

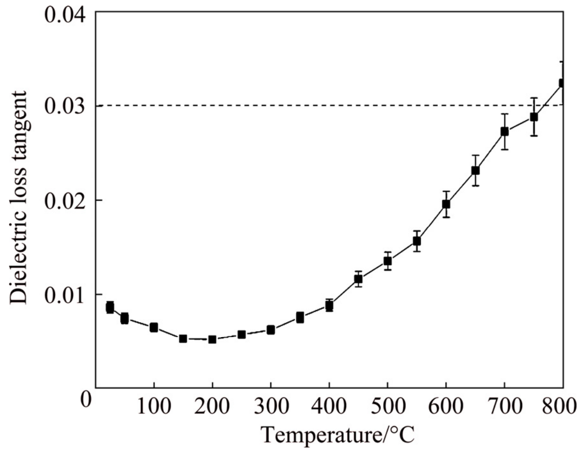

Fig. 5 Temperature dependence of dielectric loss tangent at 2.45 GHz

The reflection loss of spent automobile catalyst at different thicknesses and 2.45 GHz is shown in Fig. 6. Generally, L_R<−10 dB denotes that 68% amplitude or 90% power of electromagnetic wave is absorbed by material, while L_R<−20 dB represents that 90% amplitude or 99% power of electromagnetic wave is absorbed by material [32,33]. Obviously, the L_R of the spent automobile catalyst is temperature dependent and thickness dependent. The L_R decreased with the increase of thickness, increased from room temperature to 200 °C and decreased from 250 to 800 °C. The maximum value of L_R occurred at 200 °C. Interestingly, the value of L_R<−10 dB (−13.33 dB) was obtained only at d=0.4 m and 800 °C, indicating that the spent automobile catalyst was a medium loss material from room temperature to 750 °C. This result is similar to that of dielectric loss tangent. The peak value in Fig. 6 corresponds to the valley value in Fig. 5 at around 200 °C. Hence, the dielectric loss tangent and reflection loss are equivalent parameters to measure the microwave absorbing characteristics.

The heating curve of spent automobile catalyst (50 g) in the microwave heating equipment with power of 1000 W and frequency of 2.45 GHz is shown in Fig. 7. The temperature of spent automobile catalyst increased from 17 to 170 °C in 50 min with a heating rate of 3.06 °C/min. These

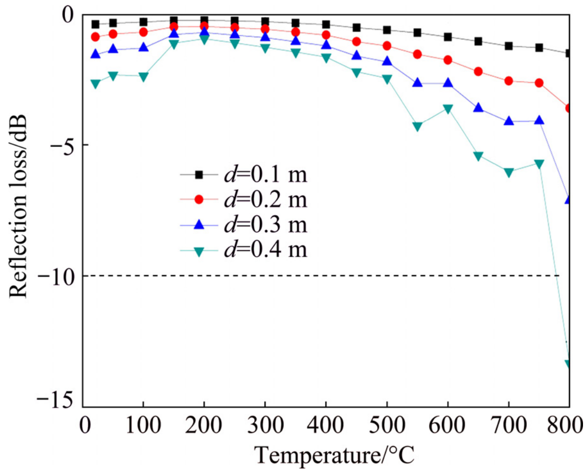

Fig. 6 Reflection loss of spent automobile catalyst at different thicknesses (d) and 2.45 GHz

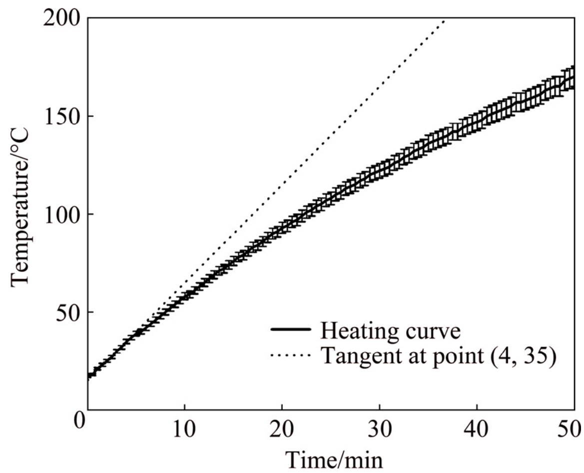

Fig. 7 Microwave heating curve of spent automobile catalyst at 2.45 GHz

experiment results showed that the spent automobile catalyst was a medium loss material. On the other hand, the temperature rise curve was gradually away from the tangent of point (4 min, 35 °C) with the increase of time, indicating that the heating rate was decreased. According to the heat conduction equation: $\rho c(\partial T/\partial t)=\nabla\cdot(k\nabla T)+1/2\omega\varepsilon_0\varepsilon''_r|\vec{E}|^2$ [34,35], the heating rate ($\partial T/\partial t$) is proportional to ε''_r at low temperatures.

The decrease of the heating rate was attributed to the decrease of the dielectric loss factor from room temperature to 170 °C, which was consistent with the data of dielectric loss factor in Fig. 4(b). Therefore, microwave heating experiment also verified that the spent automobile catalyst was medium loss material.

3.3 Average penetration depth of spent automobile catalyst

Figure 8 illustrates the effect of temperature on the calculated value and fitting curve of the power penetration depth at 2.45 GHz. The power penetration depth of the spent automobile catalyst could be calculated from Eq. (3). Obviously, the variations of the penetration depth were caused by the variations of the dielectric properties. The dielectric properties were temperature dependent, so the penetration depth was temperature dependent. The penetration depth of the spent automobile catalyst increased when the temperature increased from room temperature to 200 °C, and reached a maximum value of 2.04 m at 200 °C. When the temperature increased from 200 to 800 °C, the penetration depth decreased rapidly to 0.32 m at 800 °C. The penetration depth can be used to describe the absorbing characteristics of materials. For the spent automobile catalyst, the larger the penetration depth was, the weaker the absorbing ability was. The penetration depth is often used to determine the heating thickness of materials. However, it is only suitable for the case that the penetration depth does not change much with the increase of temperature. Hence, the average penetration depth would be employed to determine the microwave heating thickness of the spent automobile catalyst. Firstly, the variation of the penetration depth could be fitted by Gauss model as a function of temperature. The Gauss model can be expressed as exponential form [36]:

$$D_{\mathrm{p}} = D_0 + \frac{A}{w\sqrt{\pi/2}}\exp\left[-\frac{2(T-T_{\mathrm{c}})^2}{w^2}\right] \tag{4}$$

where T is the temperature; D_0, A, w and T_c are constants. The fitting results of the constants were D_0=0.3875, A=654.6956, w=334.6203, and T_c=205.3548. The coefficient of correlation (R^2) was 0.99, which implied that the fitting curve was in good agreement with the calculated value.

Secondly, the average penetration depth is determined by the following expression:

$$\bar{D}_{\mathrm{p}} = \frac{1}{T_{\mathrm{f}} - T_0}\int_{T_0}^{T_{\mathrm{f}}} D_{\mathrm{p}}(T)\mathrm{d}T \tag{5}$$

where T_0 and T_f are the initial temperature and final temperature, respectively.

Lastly, the average penetration depth of the spent automobile catalyst was $\bar{D}_{\mathrm{p}}$=1.11 m.

For microwave heating process, the average penetration depth is an intuitive physical quantity to describe the attenuation of electromagnetic energy in a material. It takes an average of 1.11 m to attenuate the microwave power density to 37% of its surface value from room temperature to 800 °C.

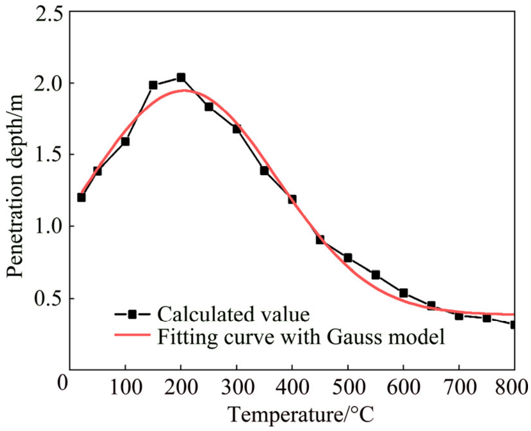

Fig. 8 Calculated value and fitting curve of power penetration depth

3.4 Thickness optimization of microwave heating for medium loss materials

Transmission line theory was employed to optimize the microwave heating thickness of a material in this study. The variation of the reflection loss with material thickness could be obtained using Mathematica 9.0 numerical calculations. Figure 9 shows the effects of the microwave heating thickness on the reflection loss at typical temperature (21, 200, 500 and 800 °C). It can be seen that the reflection loss depends on thickness and temperature. As the thickness increased, the reflection loss decreased firstly, and then increased to a constant less than −10 dB, as shown by the red arrows in Figs. 9(a, c, e, g), indicating that there was an optimum heating thickness at which almost all microwave energy was absorbed by the spent automobile catalyst. It is certainly true that the microwave energy is almost completely absorbed if the heating thickness increases excessively. For example, 90% microwave power would be absorbed by the spent automobile catalyst when the heating thickness is greater than 5 m at 21 °C in Fig. 9(a) and 1.5 m at 800 °C in Fig. 9(g). However, the larger thickness will produce cold centers. Therefore, it is necessary to optimize the microwave heating thickness of the spent automobile catalyst.

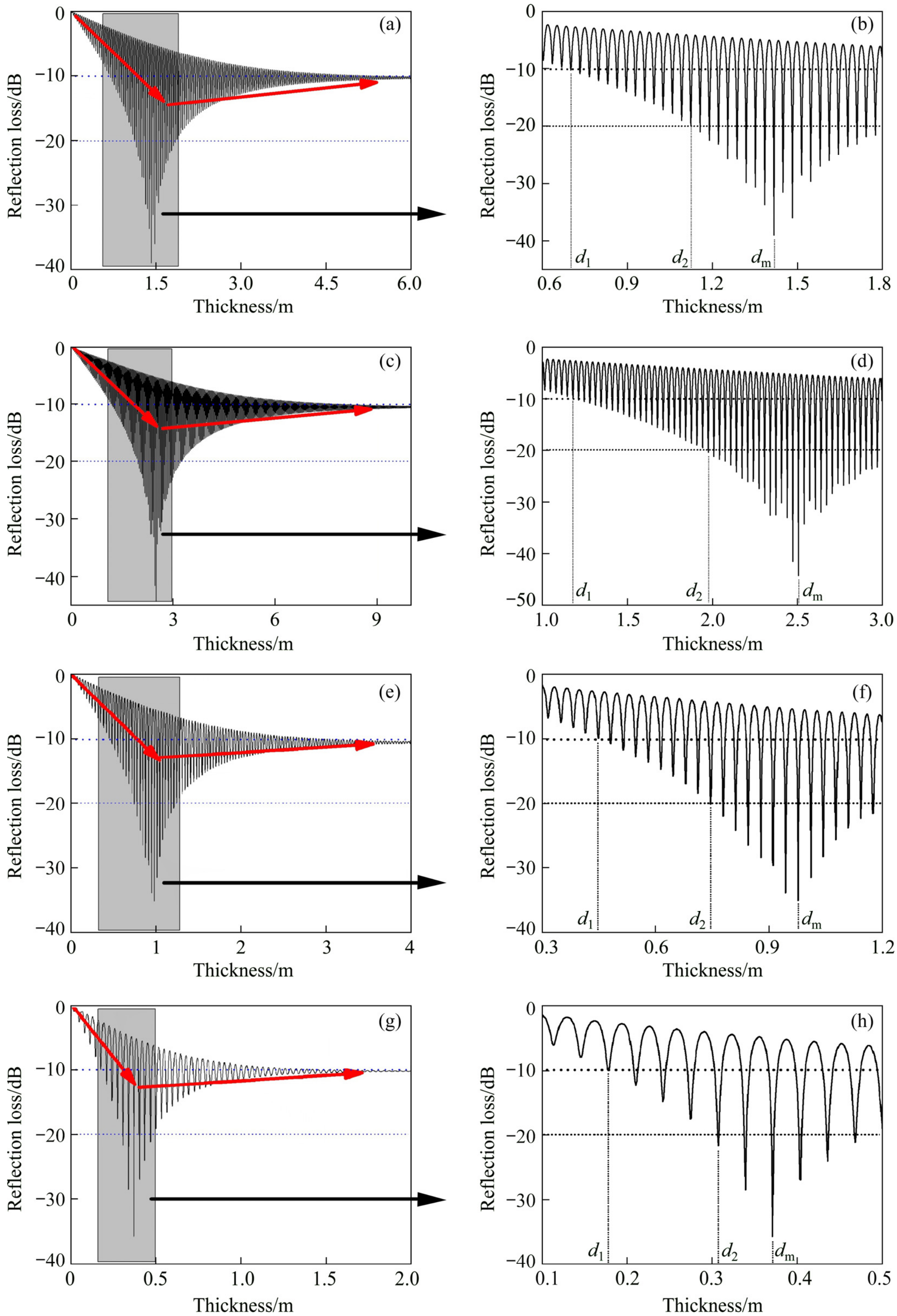

Fig. 9 Effects of microwave heating thickness on reflection loss: (a, b) 21 °C; (c, d) 200 °C; (e, f) 500 °C; (g, h) 800 °C

Here, we proposed a novel method to optimize the microwave heating thickness. In Figs. 9(b, d, f, h), d_1 is the thickness at the first valley where the reflection loss is less than −10 dB; d_2 is the thickness at the first valley where the reflection loss is less than −20 dB; d_m is the thickness at the minimum reflection loss. d_1, d_2 and d_m can be obtained by numerical calculation in the heating process. For example, d_1=0.67 m, d_2=1.24 m and d_m=1.42 m in Fig. 9(b); d_1=0.45 m, d_2=0.75 m and

d_m=0.98 m in Fig. 9(f). The values of d_1, d_2 and d_m from room temperature to 800 °C can be obtained, and their average values can also be calculated. Values of d_1, d_2 and d_m are given in Table 2. The optimum microwave heating thicknesses are about $(\bar{d}_1+\bar{d}_2)/2$ for the medium loss materials and $(\bar{d}_2+\bar{d}_m)/2$ for the high loss materials.

As discussed in Section 3.2, the spent automobile catalyst is mainly medium loss material in the studied temperature range. We can get $\bar{d}_1$=0.620 m and $\bar{d}_2$=1.048 m. Hence, we can draw a conclusion that the optimum microwave heating thickness of the spent automobile catalyst is about 0.83 m, or in form of $0.75\bar{D}_p$.

Table 2 Values of d_1, d_2 and d_m from room temperature to 800 °C

Temperature/°C	d_1/m	d_2/m	d_m/m
21	0.668	1.124	1.417
50	0.765	1.319	1.677
100	0.893	1.477	1.899
150	1.102	1.859	2.484
200	1.146	1.943	2.474
250	1.041	1.768	2.164
300	0.939	1.598	2.092
350	0.777	1.338	1.669
400	0.678	1.142	1.407
450	0.513	0.876	1.107
500	0.448	0.746	0.978
550	0.375	0.636	0.767
600	0.314	0.513	0.645
650	0.278	0.442	0.54
700	0.213	0.377	0.442
750	0.214	0.345	0.444
800	0.178	0.307	0.371

3.5 Industrial application analysis of optimum thickness

Microwave power decays exponentially as a function of distance [12,37]. According to the definition of the power penetration depth, the microwave power decreases to 37% of its surface value at penetration depth, to 14% at twice the penetration depth and to 5% at three times the penetration depth. Hence, the microwave heating thickness with 2−3 times of the penetration depth would seriously arouse the inhomogeneous distribution of temperature field.

We optimized the heating thickness of the large-scale microwave equipment. Figure 10(a) presents the photo of the large-scale microwave equipment manufactured in our laboratory. The magnetron is above the equipment, and the conveyor belt is metal. Microwave irradiates the heated materials from up to bottom. Figure 10(b) shows the schematic diagram of the optimum microwave heating chickness. The optimal thickness of $0.75\bar{D}_p$ was obtained when the microwave radiated vertically to the spent automobile catalyst. This indicated that the distance of microwave propagation was $1.5\bar{D}_p$ inside the spent automobile catalyst. Correspondingly, approximately 80% microwave power was dissipated into the optimum thickness. This conclusion takes into account a balance between the energy consumption and the uniformity of the temperature field. If microwave was oblique, it would lead to greater microwave propagation distance and then more power loss. Certainly, the higher heating rate could be obtained by using a thickness of less than $0.75\bar{D}_p$ in microwave large-scale production.

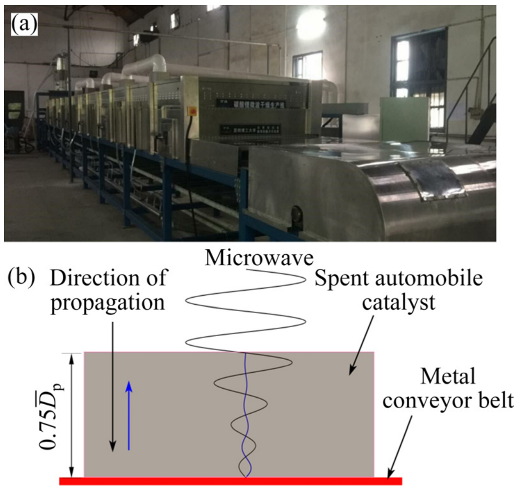

Fig. 10 Photo of large-scale microwave equipment manufactured in our laboratory (a), and schematic diagram of optimum microwave heating thickness (b)

4 Conclusions

(1) The dielectric properties of spent automobile catalyst were measured from room temperature to 800 °C at 2.45 GHZ. The microwave absorbing characteristics and heating experiment showed that spent automobile catalyst was a medium loss material.

(2) The average penetration depth ($\bar{D}_p$) calculated by Gauss model was 1.11 m.

(3) For the medium loss characteristics, the optimum microwave heating thickness was about 0.83 m or $0.75\bar{D}_p$.

(4) Industrial application analysis showed that approximately 80% of microwave power was dissipated in the optimum thickness. Moreover, the optimized method of microwave heating thickness is also suitable for the medium loss materials.

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汽车废催化剂微波加热厚度优化

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摘　要：为了提高汽车废催化剂在微波加热过程中温度场分布的均匀性，开发加热厚度优化的新方法。采用高斯模型和基于介电损耗正切和反射损耗的数值算法计算汽车废催化剂的微波加热平均穿透深度和厚度。结果表明：汽车废催化剂是一种中等损耗物料，从室温到 800 °C 的过程中平均穿透深度为 1.11 m，汽车废催化剂的最优加热厚度约为 0.83 m 或平均穿透深度的 0.75 倍。工业化应用分析显示：该最佳厚度能提高温度场的均匀分布、减少能耗。

关键词：汽车废催化剂；微波；加热厚度；优化

(Edited by Bing YANG)