Xiaobiao Shang, Junruo Chen* and Jinhui Peng

Dynamic Transmission Performances of Alumina and Mullite Refractory Ceramics in Microwave High-Temperature Heating

Abstract: This paper proposes an analytical approach to optimize the thickness of refractories for achieving maximum microwave power transmission in microwave heating based on the analysis of power transmission coefficient (PTC). The microwave PTCs of alumina (Al_2O_3) ceramics over the temperature range of 22–1,379°C at 2,450 MHz, mullite ceramics in the temperature range of 27–1,027°C at 2.45 GHz and 400–1,300°C at 915 MHz are studied. The results show that there are several transmission peaks in the PTC patterns. The transmission peak amplitude depends sensitively on the thickness of the refractory and the peak shifting towards a smaller thickness as the temperature of the refractory increases. We also show that high microwave transmission can only be achieved in a refractory with a small thickness corresponding to a slight transmission peak shift in the entire microwave heating (less than one eighth wavelength in the refractory).

Keywords: alumina, mullite, power transmission coefficient, microwave heating

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Introduction

In contrast with conventional heating, microwave heating has distinguishing characteristics, such as material-selective and volumetric heating, leading to extremely broad applications in high-temperature domain. These areas include mineral calcinations [1–3], carbothermal reduction reaction [4], ceramic sintering [5, 6]. In the field of high-temperature heating, the refractory is essential. However, the refractory used in microwave heating not only need to have basic physical and chemical properties as in conventional heating needs, but also should be a microwave transparent material or low-loss material which has better microwave transmission performance [7, 8].

Alumina and mullite ceramics have often been used to produce sample holders and lining for microwave heating due to their good fire resistance and relatively good microwave transmission at relatively low temperatures [9–15]. These applications generally require materials to withstand high temperatures under microwave irradiation. This indicates that microwave transmission capabilities of alumina and mullite ceramic at elevated temperatures are important for their utilization. In addition, the effect of the refractory thickness was generally neglected by researchers in microwave heating. The microwave transmission capability of alumina and mullite ceramics can be determined based on the analysis of power transmission coefficient (PTC) at commonly used microwave frequencies, 915 and 2,450 MHz, in a broad temperature range. To date, however, no detailed work on the PTCs of alumina and mullite ceramics at high temperatures has been reported.

To solve the above issue, our work is devoted to achieving the maximum microwave transmission in microwave high-temperature heating of materials by calculating the PTC of materials. The microwave PTCs of alumina ceramic over the temperature range of 22–1,379 °C at 2,450 MHz, the mullite in the temperature range of 27–1,027 °C at 2.45 GHz and 400–1,300 °C at 915 MHz are studied. The current work provides a general rule for thickness optimization of the refractories in microwave high-temperature heating, which could provide insight on achieving the maximum transmission during microwave high-temperature processing of materials.

Theory of PTC

Figure 1 shows the schematic diagram of the electromagnetic wave propagation in a single plate medium. PTC can be employed to quantify the amount of microwave power transmitted through the refractory layer. The larger PTC there is for a sample, the better (larger) the microwave transmits through the material. According to the transmission line theory, the theoretical PTC values of the medium can be obtained. When the wave is incident at an angle θ_i relative to the normal of the interface

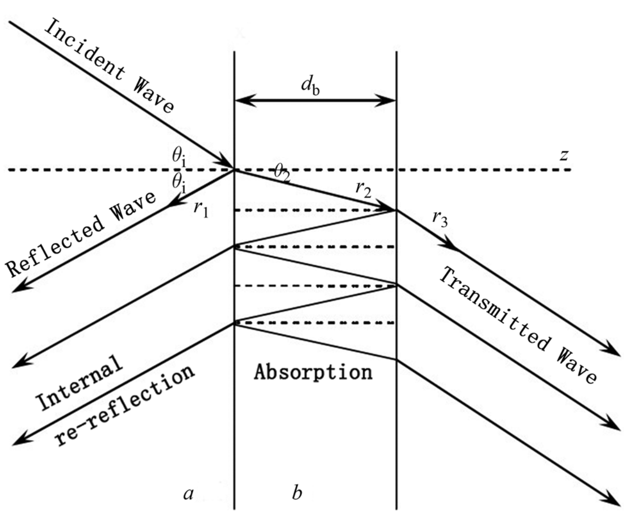

Figure 1: Schematic diagram of the electromagnetic wave propagation in a single plate medium.

between dissimilar media, the PTC is given by the following relation [16, 17]:

$$\mathrm{PTC} = t^2 \tag{1}$$

$$t = \frac{(1-{\rho_0}^2)e^{-(\alpha+j\beta)\cos(\theta_i)d_b}}{1-{\rho_0}^2 e^{-2(\alpha+j\beta)\cos(\theta_i)d_b}} \tag{2}$$

$$\rho_0 = \frac{\cos(\theta_i) - \sqrt{\varepsilon_r - \sin^2(\theta_i)}}{\cos(\theta_i) + \sqrt{\varepsilon_r - \sin^2(\theta_i)}} \tag{3}$$

$$\alpha = \frac{2\pi f}{c}\sqrt{\frac{\varepsilon'}{2}\left\{\sqrt{1+tg^2\delta}-1\right\}} \tag{4}$$

$$\beta = \frac{2\pi f}{c}\sqrt{\frac{\varepsilon'}{2}\left\{\sqrt{1+tg^2\delta}+1\right\}} \tag{5}$$

where t is the transmission coefficient, ρ_0 is a coefficient, α is the attenuation coefficient and β is the wave number of the propagating wave, ε_r is the complex relative permittivity ($\varepsilon_r = \varepsilon' - j\varepsilon''$), j is the imaginary unit, $tg\delta$ is the loss tangent ($tg\delta = \varepsilon''/\varepsilon'$), c is the velocity of light, f is the microwave frequency and d is the thickness of the medium.

From eqs (1)–(5), the PTC of refractory is determined by the relative permittivity at a given frequency as well as the thickness of the refractory. These properties of alumina and mullite ceramics are summarized in Table 1. The temperature and thickness dependences of PTC of the ceramics can be determined based on the reported parameters.

Results and discussion

The calculated results of PTC versus thickness for the alumina with increasing temperature at 2,450 MHz are shown in Figure 2. As shown in Figure 2, there are four microwave transmission peaks in the PTC patterns; it was observed that there is a matching thickness for matching

Table 1: Complex permittivities of mullite and Al_2O_3 at 915/2,450 MHz.

Parameters	Mullite[a]		Al_2O_3 (2,450 MHz)[b]				
	915 MHz ($400 \le T$°C $\le 1{,}300$)	2,450 MHz ($27 \le T$°C $\le 1{,}027$)	22 °C	491 °C	871 °C	1,050 °C	1,379 °C
ε'	$3 \cdot 10^{-9}T^3 - 4\times10^{-6}T^2 + 0.0032T + 4.4869$	$2.119\times10^{-6}T^2 - 0.000337T + 6.1438$	8.9	9.82	10.4	10.81	11.77
ε''	$9\times10^{-7}T^2 - 0.0002T - 0.0268$	$1.7052\times10^{-9}T^3 - 1.4616\times10^{-6}T^2 + 0.000559T + 0.02279$	0.004	0.025	0.093	0.158	0.476

Note: [a]Data taken from Ref. [18], [b]data taken from Ref. [19].

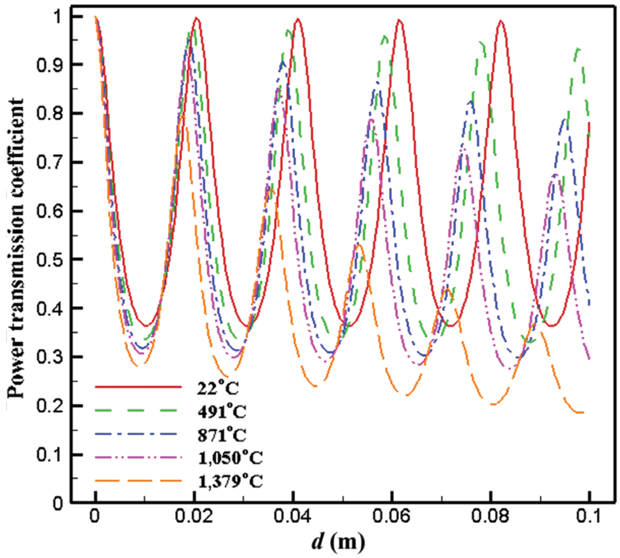

Figure 2: Temperature dependence of PTC of the Al_2O_3 slab as the thickness varies from 0 to 0.1 m at 2,450 MHz: 22 °C, 491 °C, 871 °C, 1,050 °C, 1,379 °C.

temperature at which microwave transmission is the maximum. The peak positions (d_{peak}) at various temperatures are shown in Table 2. Peak 1 at 22 °C indicates that alumina ceramic with a thickness of 20.5 mm exhibits the maximum microwave transmission (PTC = 0.9976). However, as temperature increases, a shift of peak position (Δd_{peak}) is observed. For instance, the peak shifts from 20.5 mm to 17.5 mm as the temperature increases from 22 to 1,379 °C. This indicates that a smaller thickness of the alumina ceramic is required to achieve the maximum transmission at an elevated temperature. A temperature dependence of sample thickness corresponding to the maximum microwave transmission is also observed in other peaks. This phenomenon can be attributed to the increased microwave phase constant and, therefore, a shorter microwave wavelength in alumina (λ_d) as the temperature increases (Figure 3). The value of λ_d is determined by [20]

$$\lambda_d = \sqrt{2}\lambda\left\{\varepsilon'\mu' - \varepsilon''\mu'' + \left[(\varepsilon'\mu')^2 + (\varepsilon''\mu'')^2 + (\varepsilon'\mu'')^2 + (\mu'\varepsilon'')^2\right]^{1/2}\right\}^{-1/2} \quad (6)$$

where λ is the microwave wavelength in free space.

In Figure 2 it can be further noticed that for all transmission peaks (peaks 1–4) in the PTC patterns, the amplitude of peak decreases with increasing temperature. For example, PTC of peak 1 decreases from 0.9976 to 0.7969 as the

Table 2: Transmission peak positions in the PTC of mullite and Al_2O_3 ceramics at various temperatures.

Peak nos.												Mullite (2,450 MHz)
	27 °C		200 °C		400 °C		600 °C		800 °C		1,000 °C	
	d_{peak} (mm)	PTC	d_{peak} (mm)	PTC	d_{peak} (mm)	PTC	d_{peak} (mm)	PTC	d_{peak} (mm)	PTC	d_{peak} (mm)	PTC
1	24.5	0.9730	24.5	0.9370	24.0	0.9171	23.5	0.8728	22.5	0.7727	21.0	0.6249
2	49.5	0.9472	49.0	0.8781	48.5	0.8426	47.0	0.7648	45.0	0.6053	42.5	0.4058
3	74.0	0.9225	74.0	0.8242	72.5	0.7750	70.5	0.6725	67.5	0.4798	64.0	0.2725
4	99.0	0.8977	98.5	0.7746	97.0	0.7147	94.0	0.5934	90.5	0.3844	85.0	0.1874
												Mullite (915 MHz)
	400 °C		600 °C		800 °C		1,000 °C		1,200 °C		1,300 °C	
	d_{peak} (mm)	PTC	d_{peak} (mm)	PTC	d_{peak} (mm)	PTC	d_{peak} (mm)	PTC	d_{peak} (mm)	PTC	d_{peak} (mm)	PTC
1	71.0	0.9705	68.5	0.8739	65.0	0.7599	60.5	0.6506	55.5	0.5599	53.0	0.5229
												Al_2O_3 (2,450 MHz)
			22 °C		491 °C		871 °C		1,050 °C		1,379 °C	
			d_{peak} (mm)	PTC	d_{peak} (mm)	PTC	d_{peak} (mm)	PTC	d_{peak} (mm)	PTC	d_{peak} (mm)	PTC
1			20.5	0.9976	19.5	0.9863	19.0	0.9519	18.5	0.9220	17.5	0.7969
2			41.0	0.9953	39.0	0.9727	38.0	0.9068	37.0	0.8506	35.5	0.6462
3			61.5	0.9928	58.5	0.9592	57.0	0.8644	56.0	0.7870	53.5	0.5292
4			82.0	0.9904	78.0	0.9459	76.0	0.8247	74.5	0.7312	71.0	0.4388
5			>100	–	97.5	0.9327	95.0	0.7873	93.0	0.6795	89.0	0.3671

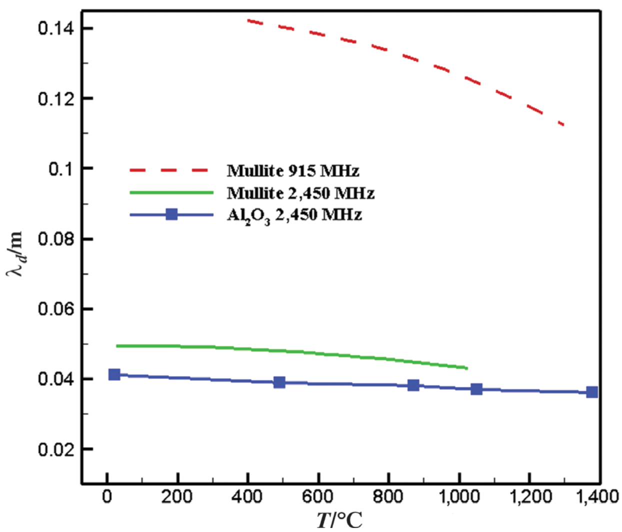

Figure 3: Temperature dependence of the microwave wavelength in mullite and Al_2O_3 ceramics.

temperature increases from 22 to 1,379 °C. This can be explained by the fact that microwave penetration depth (D_p) of the alumina decreases with increasing temperature (Figure 4). The value of D_p is determined by [21]

$$D_p = \frac{\lambda}{2\sqrt{2}\pi}\left\{\varepsilon''\mu'' - \varepsilon'\mu' + \left[(\varepsilon'\mu')^2 + (\varepsilon''\mu'')^2 + (\varepsilon'\mu'')^2 + (\mu'\varepsilon'')^2\right]^{1/2}\right\}^{-1/2} \quad (7)$$

These suggest that the highest microwave transmission cannot be achieved throughout the microwave heating in the refractories with a fixed thickness because of the temperature dependence of the variation of peak amplitude and the shift of microwave transmission peaks. However, we can anticipate that a suitable thickness should be limited in the range indicated by the positions of the microwave transmission peaks in the temperature range (i.e., $d_{\text{peak},1375^\circ\text{C}} \le d \le d_{\text{peak},22^\circ\text{C}}$). This suggests that thicknesses for the alumina ceramic corresponding to four peaks in the PTC patterns, as shown in Figure 5, should be chosen for achieving high transmission throughout the microwave heating process.

It is also seen from Figure 2 that the Δd_{peak} becomes larger with the number of peak corresponding to larger thickness. This suggests that the shift of peak is slight at small thicknesses and becomes large with increasing thickness; therefore, it is impossible to achieve the highest PTC with larger shift of transmission peak.

Figure 5 shows the temperature dependence of PTC of the alumina as the thickness varies from 19 mm to 76 mm. It is seen from Figure 5 that the alumina ceramic has the maximum transmission ($0.7212 \le \text{PTC} \le 0.9128$) when the alumina ceramic has a thickness of 19 mm which corresponds to peak 1 in Figure 2. This PTC range indicates that more than 70% power is transmitted throughout the microwave heating process. The high-power transmission is essentially attributed to a small peak shift in the temperature range ($\Delta d_{\text{peak}} = d_{\text{peak},22^\circ\text{C}} - d_{\text{peak},1379^\circ\text{C}} \le \lambda_d/8$). For the PTC curve of alumina with a larger thickness, microwave transmission tends to decrease. This is particularly true for the alumina having a thickness greater than 76 mm due to a significant variation of PTC with temperature.

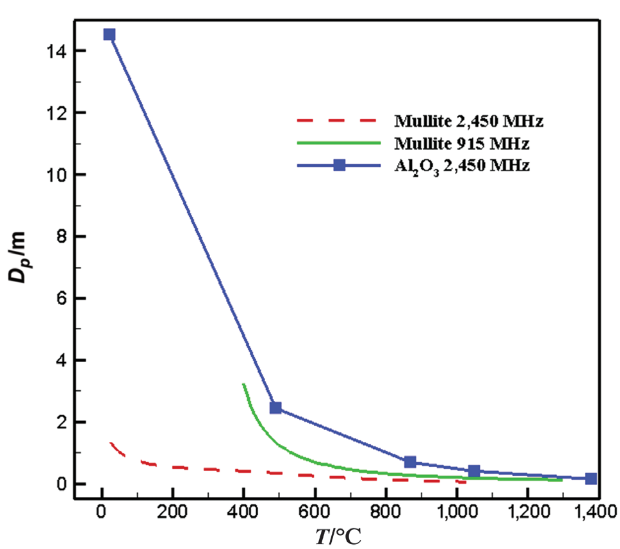

Figure 4: Temperature dependence of the microwave penetration depth of mullite and Al_2O_3 ceramics.

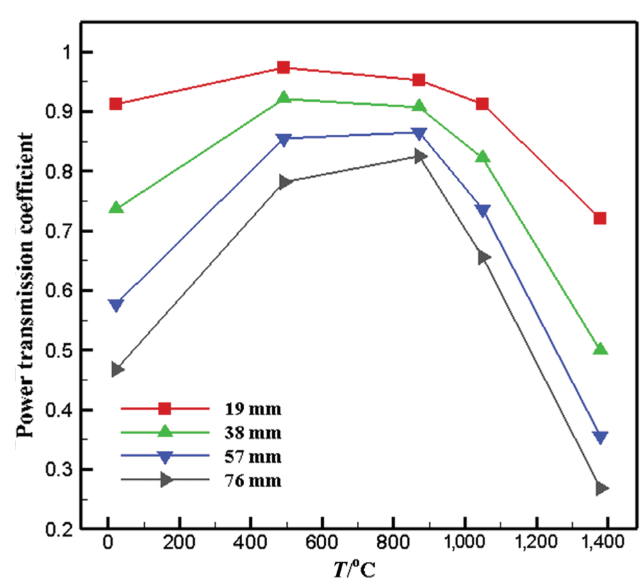

Figure 5: Temperature dependence of PTC of the Al_2O_3 slab as the thickness varies from 19 mm to 76 mm at 2,450 MHz.

The calculated results of PTC versus thickness for mullite with increasing temperature at 2,450 MHz are shown in Figure 6. As shown in Figure 6, there are also four microwave transmission peaks in the PTC patterns, which can be attributed to the microwave resonance. The peak positions (d_{peak}) at various temperature are shown in Table 2. Peak 1 at 27 °C indicates that the mullite ceramic with a thickness of 24.5 mm exhibits the maximum microwave transmission (PTC = 0.9730). However, as temperature increases, a shift of peak position (Δd_{peak}) is observed. For instance, the peak shifts from 24.5 mm to 21 mm as the temperature increases from 27 to 1,000 °C. A temperature dependence of sample thickness corresponding to the maximum microwave transmission is also observed in other peaks. This phenomenon can also be attributed to the increased microwave phase constant and, therefore, a shorter microwave wavelength in mullite (λ_d) as the temperature increases (Figure 3).

It is also seen from Figure 6 that Δd_{peak} becomes larger with the peak number increasing. This suggests that the shift of peak is slight at small thicknesses and becomes large with the increasing thickness which occurs in the PTC patterns of the alumina; therefore, it is impossible to achieve the highest PTC with larger shift of transmission peak throughout the microwave heating process.

In Figure 6 it can be further noticed that for all transmission peaks (peaks 1–4) in the PTC patterns, the amplitude of peak decreases with increasing temperature. For example, the PTC of peak 1 decreases from 0.9730 to 0.6249 as the temperature increases from 27 to 1,000 °C. This can also be explained by the fact that microwave penetration depth (D_p) of the alumina decreases with increasing temperature (Figure 4).

The calculated results of PTC versus thickness for mullite with increasing temperature at 915 MHz are shown in Figure 7. As shown in Figure 7, there is only one microwave transmission peak in the PTC patterns. The

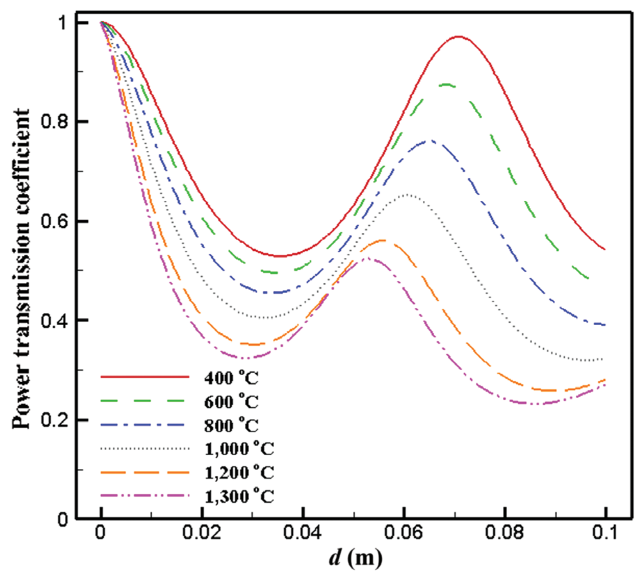

Figure 7: Temperature dependence of PTC of the mullite slab as the thickness varies from 0 to 0.1 m at 915 MHz: 400 °C, 600 °C, 800 °C, 1,000 °C, 1,200 °C, 1,300 °C.

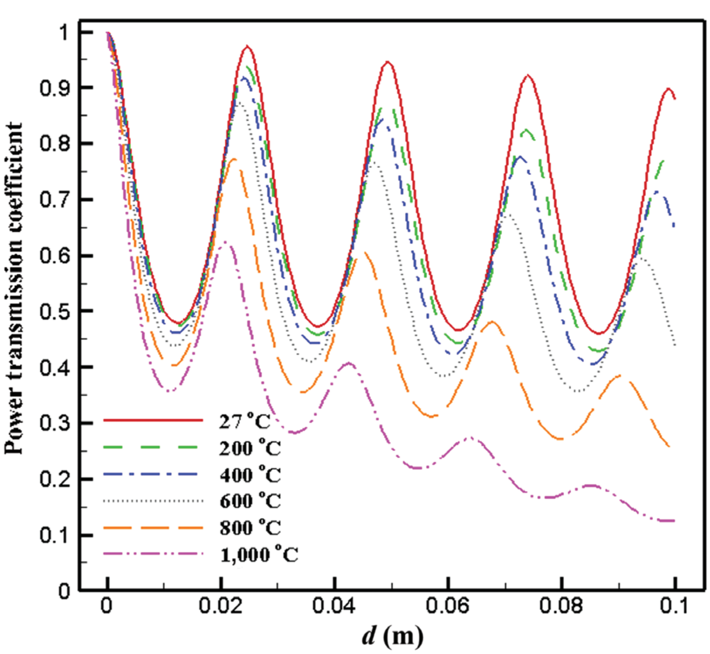

Figure 6: Temperature dependence of PTC of the mullite slab as the thickness varies from 0 to 0.1 m at 2,450 MHz: 27 °C, 200 °C, 400 °C, 800 °C, 1,000 °C.

phenomenon can be explained by a shorter microwave wavelength in mullite at 915 MHz. The peak positions (d_{peak}) at various temperatures are shown in Table 2. Peak 1 at 400 °C indicates that mullite ceramic with a thickness of 71 mm exhibits the maximum microwave transmission (PTC = 0.9705). However, as temperature increases, a shift of peak position (Δd_{peak}) is observed. For instance, the peak shifts from 71 mm to 53 mm as the temperature increases from 400 to 1,300 °C. This phenomenon can also be attributed to the increased microwave phase constant and, therefore, a shorter microwave wavelength in mullite (λ_d) as the temperature increases (Figure 3).

In Figure 7 it can be further noticed that for all transmission peaks (peaks 1–4) in the PTC patterns, the amplitude of peak decreases with increasing temperature. For example, PTC of the peak decreases from 0.9705 to 0.5229 as the temperature increases from 400 to 1,300 °C. This can also be explained by the fact that microwave penetration depth (D_p) of mullite decreases with increasing temperature (Figure 5).

The PTC value of ≥ 0.7 is comparable to the 70% of microwave transmission and thus "PTC ≥ 0.7" is considered as an adequate microwave transmission [22, 23]. PTC values more than 0.7 are recorded, as shown in Table 3.

Conclusions

The analytical approach is proposed to optimize the thickness of the refractories for achieving maximum microwave power transmission in microwave heating based on the analysis of PTC. The microwave PTCs of the alumina ceramic over the temperature range of 22–1,379 °C at 2,450 MHz, the mullite in the temperature range of 27–1,027 °C at 2.45 GHz and 400–1,300 °C at 915 MHz were studied. The calculated results show that the microwave PTC depends sensitively on the thickness of the refractories, and there is a set of transmission peaks in the PTC patterns of alumina and mullite ceramics, respectively. The transmission peak shifts towards a smaller thickness as the temperature of the ceramic increases, and Δd_{peak} becomes larger with the peak number increasing. We also show that high microwave transmission can only be achieved in a refractory with a small thickness corresponding to a slight transmission peak shift in the entire microwave heating (less than one eighth wavelength in the refractory). The analytical approach presented in this paper should be useful in selecting the refractories and designing microwave applicators.

Table 3: The thickness ranges of the power transmission coefficient more than 70%.

Peak nos.	Mullite											
	2,450 MHz						915 MHz					
	27 °C	200 °C	400 °C	600 °C	800 °C	1,000 °C	400°C	600 °C	800 °C	1,000°C	1,200 °C	1,300 °C
	Thickness (mm)	Thickness (mm)	Thickness (mm)	Thickness (mm)	Thickness (mm)	Thickness (mm)	Thickness (mm)	Thickness (mm)	Thickness (mm)	Thickness (mm)	Thickness (mm)	Thickness (mm)
1	0–5	0–5	0–4.5	0–4.5	0–3.5	0–3	0–17	0–15	0–12	0–10	0–8	
2	19.5–29.5	20–29	20–28.5	20–27	20–24.5		54.5–87.5	55.5–80.5	58.0–71.5			
3	44.5–54	45–53	45–52	45–49								
4	69.5–78.5	70–77	70–75									
5	94.5–100	96–100	96–98									

Peak nos.	Al_2O_3 (2,450 MHz)				
	22 °C	491 °C	871 °C	1,050 °C	1,379 °C
	Thickness (mm)	Thickness (mm)	Thickness (mm)	Thickness (mm)	Thickness (mm)
1	0–3	0–2.5	0–2.5	0–2.5	0–2
2	17.5–23.5	17–22	16.5–21.5	16.5–20.5	16.5–19
3	38–44	36.5–41.5	35.5–40	35.5–39	
4	58.5–64.5	56–61	55–59	54.5–57	
5	79–85	75.5–80.5	74–77.5	73.5–75	
6	99.5–100	95–100	93.5–96.5		

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