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The Numerical Analysis of Non-polar Solvent Heated by Microwave Indirectly

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When the dielectric properties of the sample are too poor to allow efficient heating by microwave radiation, the use of container that have large loss tangent values can significantly overcome these problems and enable adequate heating of the whole sample. In this paper, carbon tetrachloride at silicon carbide container with three different configurations were heated indirectly by microwave. The uniformities of temperature were analyzed using COMSOL Multiphysics under different configurations during microwave heating. It was concluded that the temperature uniformities were influenced by the parameters of different configurations, when the improvement in electric field uniformity is not obvious for one configuration, it can be significantly improved by changing to another specific configuration.

Key Words: Microwave heating, Non-polar solvent, Temperature uniformity, Multiphysics.

INTRODUCTION

In the early 1980's, microwaves were proposed to be used for accelerating chemical reactions by their efficient heating of some reactants. Many recent reports indicated that microwave could evidently accelerate reactions, and the rate enhancement factor could reach over one thousand. Most chemical reactions are sensitive to temperature therefore using microwaves to heat reactants presents an impressive application prospect[1,2]. At present, in the reactions assisted and catalyzed, the majority of reaction solvents are polar in order that they are heated preferably. However, the non-polar solvents are also commonly used in a number of chemical reactions.

When the dielectric properties of the sample are too poor to allow efficient heating by microwave radiation, the use of container that have large loss tangent values can significantly overcome this problem and enable adequate heating of the whole sample. This often provides an efficient way of using non-polar solvents for running chemical syntheses using microwave radiation. However, this method might cause heterogeneous temperature distribution in chemical syntheses. Thus, it is necessary to analyze some parameters influencing temperature uniformities using numerical calculation.

Due to the high microwave absorptivity, with regard to silicon carbide, materials (*i.e.* a solvent or a reaction mixture) contained inside the container will be effectively shielded from the electromagnetic field[3]. The heat flow through the walls of the reaction vessel and subsequent heat exchange with the reaction mixture contained inside is exceptionally fast based on high thermal conductivity and diffusibility of silicon carbide[4].

Carbon tetrachloride is a non-polar solvent widely used in synthetic chemistry research. This paper therefore focused on the heating of carbon tetrachloride in silicon carbide container with different constructions. The uniformity of temperature is studied during heating. This study will provide further assists and references for the system of microwave promoting non-polar solvent reaction.

Besides, there is a vital problem for microwave heating that the dielectric heating process represents a coupled electrothermal problem. According to the classification made by Kumbhar and Kulkarni[5] the coupling of electromagnetic and thermal fields is week, due to the large difference in the time constants of the two problems. Thus it is necessary to consider the two problems which are the results of the electric field analysis allowing the determination of the source term for the heat diffusion equation and the analysis of the temperature distribution in the load enabling a re-evaluation of the physical constants (permittivity, loss angle, *etc.*), which in turn will modify the electric field distribution[6]. COMSOL can solve effectively the coupled electrothermal problem and it is chosen finally for this study.

METHODS AND SIMULATIONS

In this paper, the COMSOL software based on FEM is used to analyze the uniformity of electric field and temperature.

Three configurations are separately presented to measure the uniformity of the electric field and temperature in the non-solvent. Their physical models are showed in Figs. 1-3.

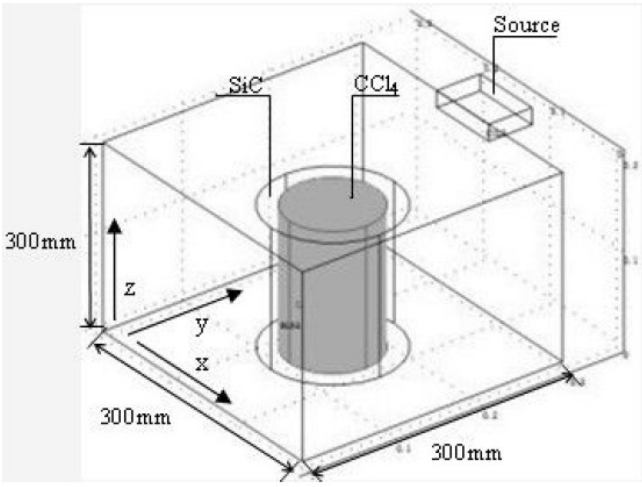


Fig. 1. Inside material is CCl_4 and the outer is silicon carbide

Configuration 1: The problem to be solved consists of a 300 × 300 × 300 mm^3 metallic rectangular applicator. In this model, the inside material is CCl_4 and the outer is silicon carbide. The cubage of CCl_4 is $V_{CCL4} = \pi^* r^{2*} h = \pi^* 0.03^{2*}0.15 \approx 4.192869e\text{-}4\ (m^3)$ and the cubage of SiC is $V_{SIC} = \pi^* R^{2*} h\text{-}\pi^* r^{2*} h = \pi^* 0.15^* (0.0424^{2\text{-}}0.03^2) \approx 4.190336e\text{-}4\ (m^3)$. R is the radius of big cylinder, r is the radius of small cylinder, and h is the height of carbon tetrachloride and silicon carbide.

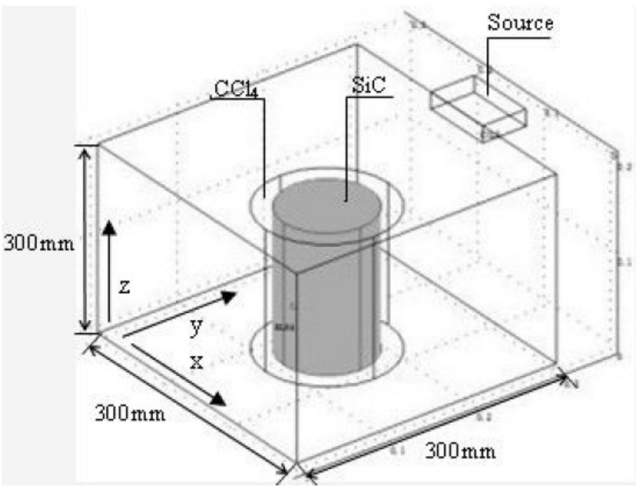


Fig. 2. Inside material is silicon carbide and the outer is CCl_4

Configuration 2: The problem to be solved consists of a 300 × 300 × 300 mm^3 metallic rectangular applicator. In this model, the inside material is silicon carbide and the outer is CCl_4. The cubage of SiC is $V_{SIC} = \pi^* r^{2*} h = \pi^* 0.03^{2*} 0.15 \approx 4.192869^{-4}\ (m^3)$ and the cubage of CCl_4 is $V_{CCL4} = \pi^* R^{2*} h\text{-}\pi^* r^{2*} h = \pi^* 0.15^* (0.0424^{2\text{-}}\ 0.03^2) \approx 4.190336^{-4}\ (m^3)$. R is the radius of big cylinder, r is the radius of small and h is the common height of CCl_4 and silicon carbide.

Configuration 3: The problem to be solved consists of a 300 × 300 × 300 mm^3 metallic rectangular applicator. In this model, the outer and inside material is silicon carbide and the middle is CCl_4. The cubage of CCl_4 is $V_{CCL4} = \pi^*R^{2*}h\text{-}\pi^* r^{2*}h = \pi^* (0.0316^{2\text{-}}0.01^2)\ ^*0.15 \approx 4.235949e\text{-}4\ (m^3)$ and the cubage of SiC is $V_{SIC} = \pi^*R'^{2*}h\text{-}\pi^*R^{2*}h = \pi^*r^{2*}\ h = \pi^*\ (0.0424^{2\text{-}}\ 0.0316^2 + 0.012)\ ^*0.15 \approx 4.243118e\text{-}4\ (m^3)$. R is the radius of big cylinder and R' is the radius of middle cylinder, r is the radius of small cylinder and h is the common height of CCl_4 and silicon carbide.

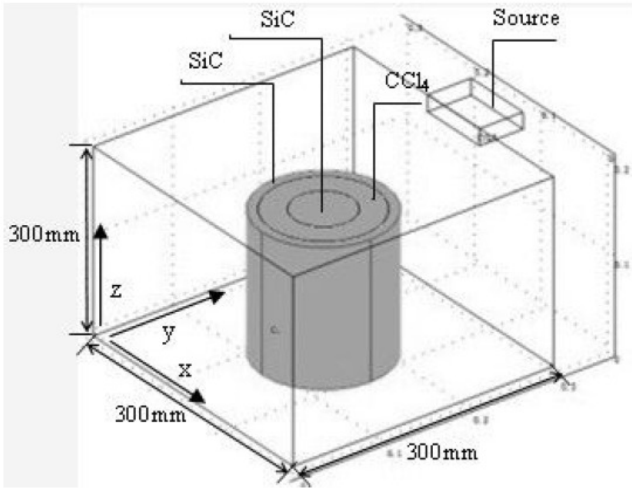


Fig. 3. Outer and inside material is silicon carbide and the middle is CCl_4

The permittivity of CCl_4 was taken from reference[7], which is $\varepsilon_r = 2.24\text{-}j^*\ 0.002$. The permittivity of silicon carbide changes with temperatures, so it is necessary to establish a function between the permittivity and temperature. From previous works[4], the relation between tan δ and temperature is gained as Fig. 4.

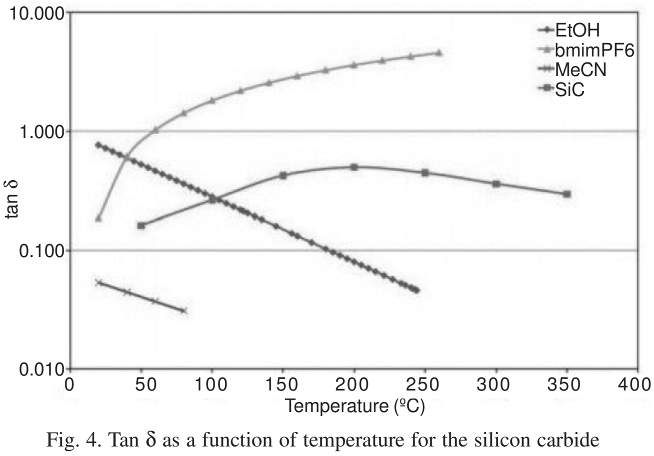


Fig. 4. Tan δ as a function of temperature for the silicon carbide

The values of four special points from the curve of silicon carbide are analyzed in Table-1.

TABLE-1
LOSS TANGENT FROM THE SILICON CARBIDE CURVE

T (ºC)	50	100	150	200
Tan δ	0.18	0.33	0.60	0.69

According to the definition of loss tangent, tan δ is imaginary part dividing real part of the permittivity. It is assumed $\varepsilon_r = a\text{-}j^*b$, so tan δ = b/a. The real part of silicon carbide permittivity, which is obtained from literature is a = 6.52 and its imaginary part which is obtained by linear fit (Table-1) is b = -6.40786 + 0.02347* T. Finally, the permittivity of SiC is ε_r = 6.52-j* (-6.40786 + 0.02347* T). During all the computational process, the input power at the waveguide feeding has been set to P = 700 W. The microwave heating time is t = 5s. Relative parameters which are used to solve the case are listed in Table-2.

TABLE-2
RELATIVE PARAMETERS

Parameter	SiC	CCl_4
Heat conduction ; k (w/m*K)	20	0.106
Density : σ(kg/m^3)	3200	1595
Atmospheric heat capacity : C_p(J/kg* ºC)	170	854

After calculation, the results such as maximum and minimum value of electric field and temperature were recorded during heating. Then the uniformities of electric field and temperature were obtained respectively by formula (1) and (2). The average temperature was achieved by formula (3).

Finally, comparing synthetically the configuration1, 2 and 3, the uniformity of electric field and temperature is gained.

$$\text{E-field uniformity} = \sqrt{\frac{|E_{max}| - |E_{min}|}{|E_{max}|}} \quad (1)$$

where, E_{max} is the maximum electric field and E_{min} is minimum electric field.

$$\text{Temperature uniformity} = \sqrt{\frac{T_{max} - T_{min}}{T_{max}}} \quad (2)$$

where, T_{max} is the maximum temperature and T_{min} is minimum temperature.

$$\text{Average temperature} = \frac{\sum_{i=1}^{\infty} T_i}{V} \quad (3)$$

where, T_i is the temperature for every grid of heated materials.

RESULTS AND DISCUSSION

The three different configurations were calculated and the following research reports in detail their results during heating.

Electric field and temperature distribution of CCl_4

Configuration 1

Electric field distribution: Fig. 5 shows the electric field distribution of X = 0.15 m section. It is observed that the upper section (near the source) is intense and the middle is poor relatively. And the electric field of silicon carbide is poor.

From Fig. 6, the field intensity is decreasing from inside to outside. The maximum lies in the center, and the electric field of silicon carbide is poor as well.

Temperature distribution: It is observed from Fig. 7 that the variation of the temperature for the heated CCl_4 is much smaller and almost keeps at 325 K. However, the temperature of silicon carbide is reducing gradually.

Fig. 8 shows the temperature distribution of Z = 0.1 m section. It is observed that the uniformity of the temperature for heated CCl_4 is better and the maximum value almost locates in the source.

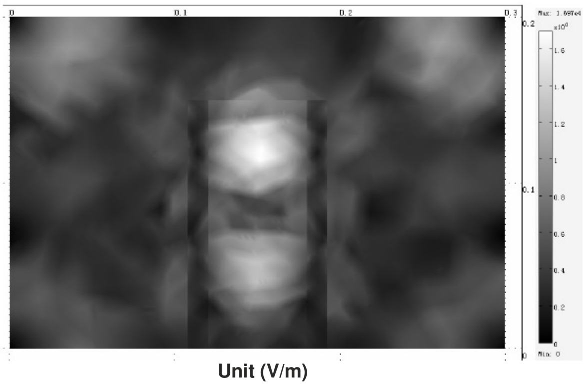


Fig. 5. Electric field distribution X = 0.15 m

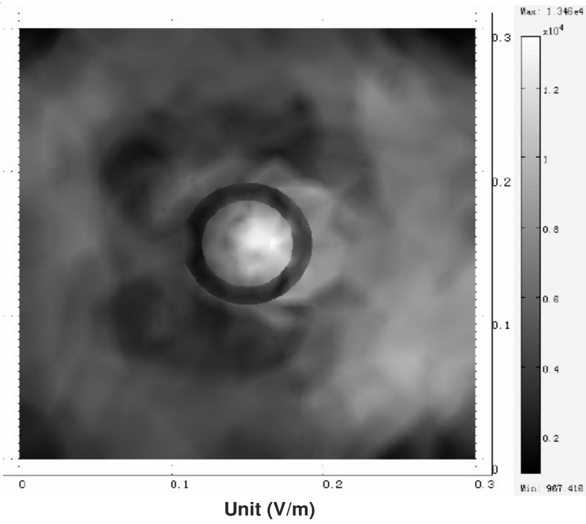


Fig. 6. Electric field distribution Z = 0.1 m

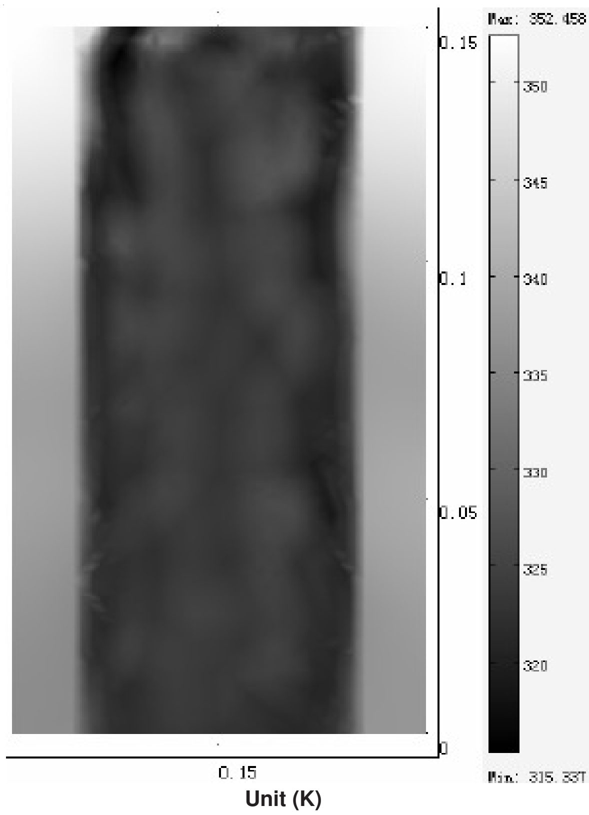
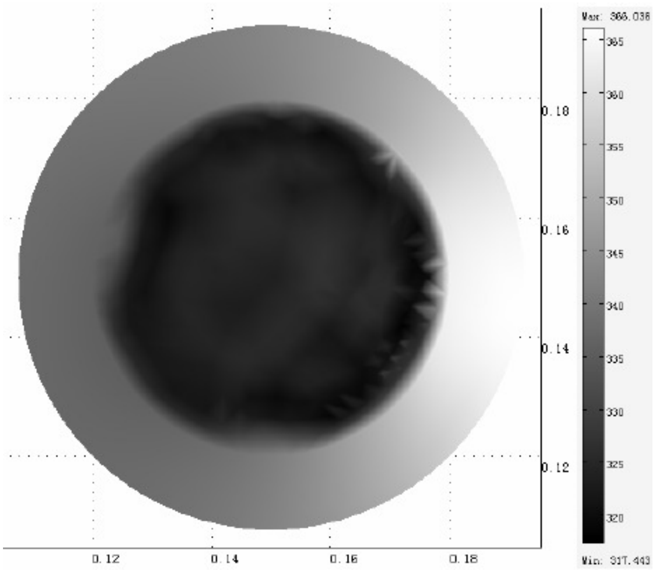


Fig. 7. Temperature distribution X = 0.15 m

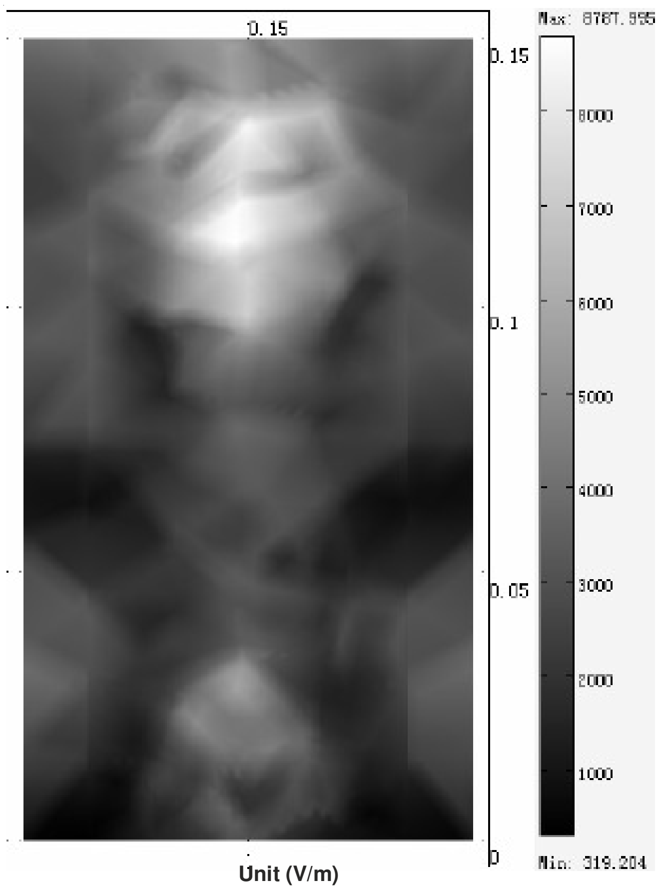


Unit (K)

Fig. 8. Temperature distribution Z = 0.1 m

Configuration 2

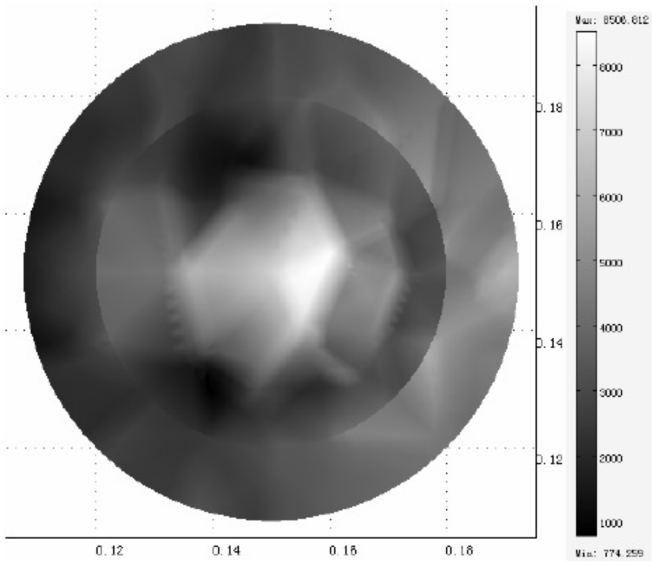
Electric field distribution: Fig. 9 shows the electric field distribution of X = 0.15 m section. It is observed that the electricfield distribution is relatively uniform at the bottom of CCl_4. However, the field of the upper silicon carbide is strong and the lower is weak, which is thought to be due to the result of the strong microwave absorption for silicon carbide.



Unit (V/m)

Fig. 9. Electric field distribution X = 0.15 m

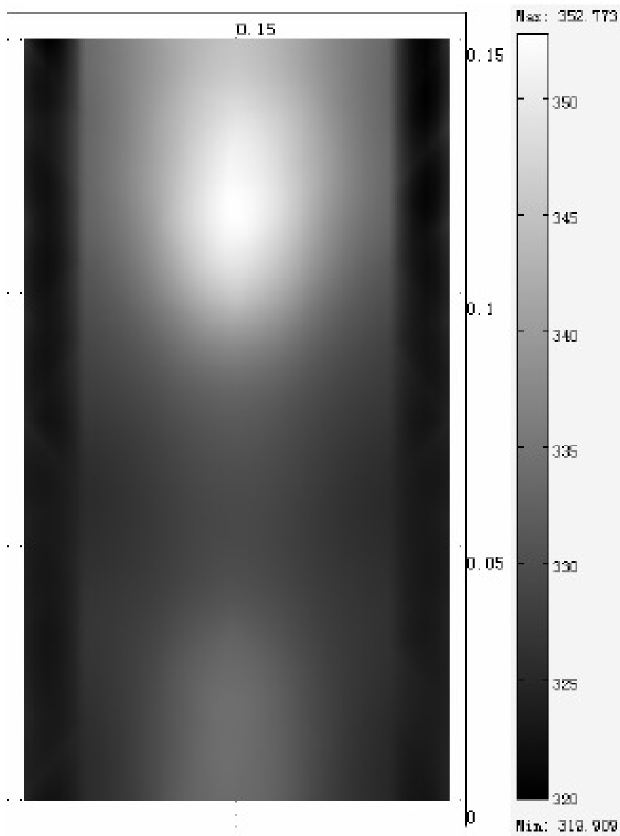
Fig.10 shows the electric field distribution of Z = 0.1 m section. It is observed that the electric field in silicon carbide is strong.



Unit (V/m)

Fig. 10. Electric field distribution Z = 0.1 m

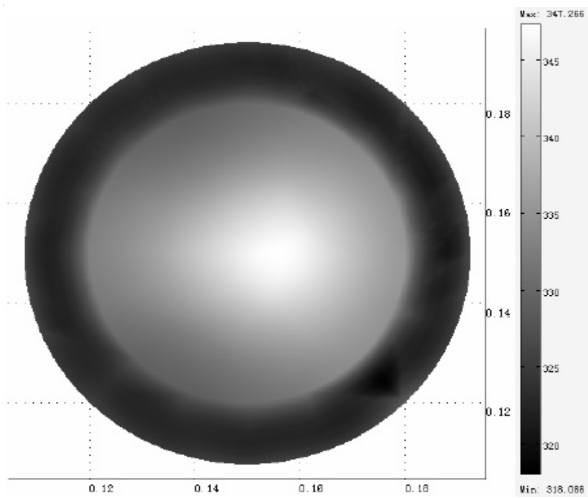
Temperature distribution: Fig. 11 shows the temperature distribution of X = 0.15 m section. It is observed that the temperature of the heated CCl_4 increases gradually from top to bottom over vertical section and the maximum value presents at the bottom.



Unit (K)

Fig. 11. Temperature distribution X = 0.15 m

Fig. 12 shows the temperature distribution of Z = 0.1 m section. It is observed that the temperature is almost uniform over the horizontal section and maximum value locates in the source. Besides, the temperature of inside silicon carbide is high.

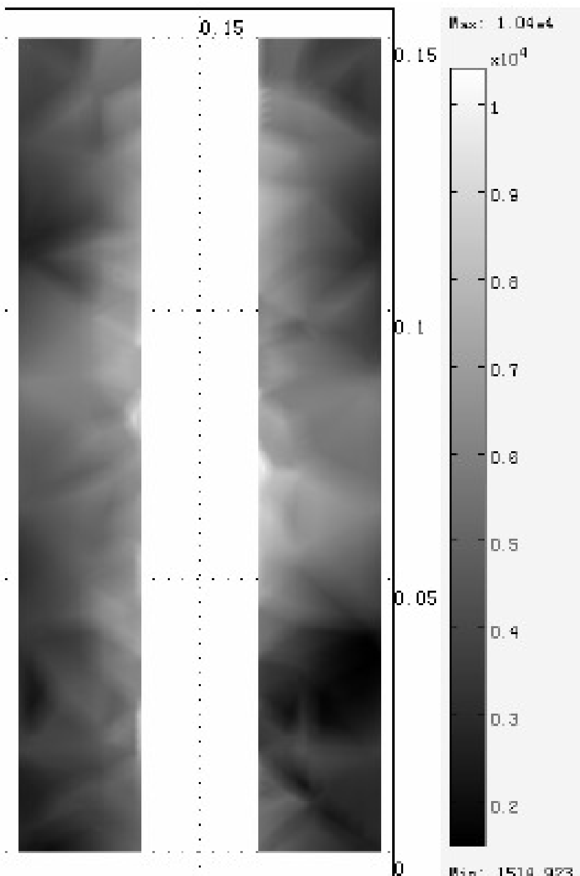


Unit (K)

Fig. 12. Temperature distribution Z = 0.1 m

Configuration 3

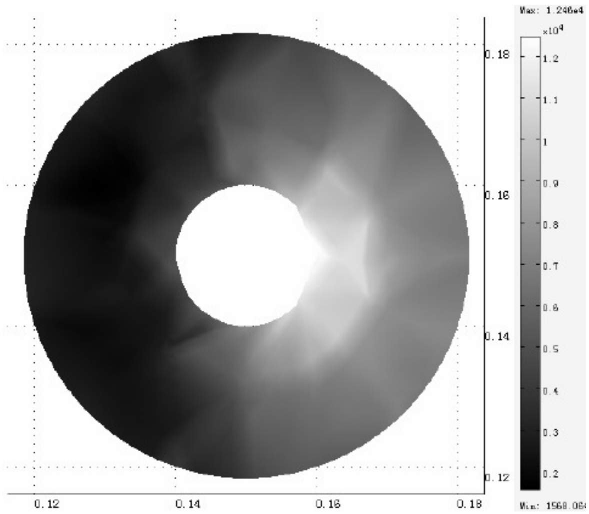
Electric field distribution: Fig. 13 shows the electric field distribution of X = 0.15 m section. It is observed that the electric field of squeezed CCl_4 is uniform relatively.



Unit (V/m)

Fig. 13. Electric field distribution X = 0.15 m

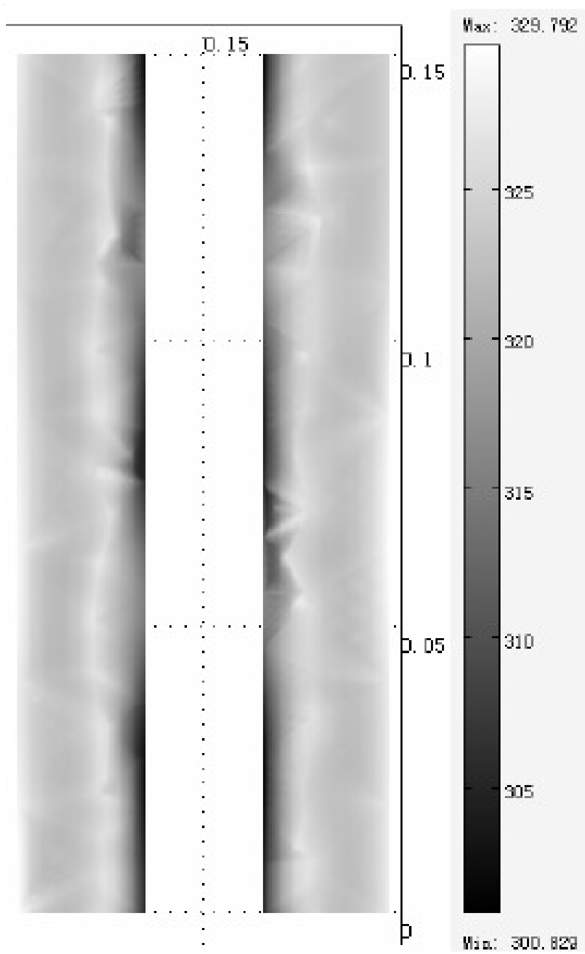
Fig. 14 shows the electric field distribution of Z = 0.1 m section. It is observed that the electric field of CCl_4 is high near the source.



Unit (V/m)

Fig. 14. Electric field distribution Z = 0.1 m

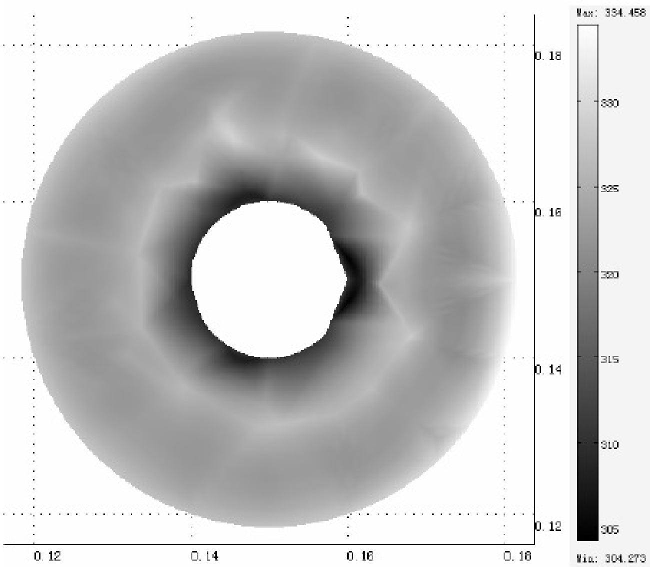
Temperature distribution: Fig.15 shows the temperature distribution of X = 0.15 m section. It is observed that the temperature of CCl_4 almost reaches a steady value.



Unit (K)

Fig. 15. Temperature distribution X = 0.15 m

Fig. 16 shows the temperature distribution of Z = 0.1 m section. It is observed that the heated effect of CCl_4 is obvious on the outside and the temperature is high relatively.



Unit (K)

Fig. 16. Temperature distribution Z = 0.1 m

Analysis and comparison: According to simulation results, the maximum and minimum value of the temperature and the electric field were recorded during the microwave heating process. Finally, the uniformity of the electric field and temperature were calculated by the formula (1) and (2). The results from three configurations for the electric field and temperature are listed in Table-3.

TABLE-3
RESULTS OF THE THREE CONFIGURATIONS

CCl_4 solvent	Config. 1	Config. 2	Config. 3
E_{max} (V/m)	18148.451	10194.867	42896.772
E_{min} (V/m)	175.316	89.029	131.163
E-field uniformity	0.99516	0.9956	0.99847
T_{max} (ºC)	384.08	348.431	340.983
T_{min} (ºC)	297.968	312.704	300.704
Temperature uniformity	0.4735	0.3242	0.3437
Average temp.: T_{avg} (ºC/m^3)	325.1846	333.2234	322.925

By comparing the data in Figs. 5-16 and Table-3, one can observe that configuration 1 is superior to configuration 2, with configuration 3 the worst for the electric field uniformity. For temperature uniformity, configuration 2 is superior to configuration 3 and configuration 3 is superior to configuration 1. For average temperature, configuration 2 is superior to configuration 1 and configuration 1 is superior to configuration 3. To sum up, configuration 2 is considered synthetically as the best configuration among the three configurations. It is the configuration that silicon carbide is surrounded by CCl_4.

From above, it is obvious that the temperature uniformities were influenced by different configurations, the uniformity of temperature can be highly improved by changing the configuration.

Conclusion

The microwave absorption of the non-polar solvent is poor and cannot be heated by microwave radiation. This paper used silicon carbide with the high microwave absorption properties as the heated container and CCl_4 as the non-polar solvent. Three configurations were designed to heat CCl_4 assisted by SiC. Best configuration with uniformity of electric and temperature field was achieved. It is the configuration that SiC is surrounded by CCl_4.

By calculation, the three configurations are integrally compared in the uniformity of electric and temperature field. When the improvement in electric field uniformity is not obvious, it can improve significantly the uniformity of temperature by changing the configuration.

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