Temperature Dependence of Microwave Dielectric Properties of SiO_2 /BN Composite

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Abstract. In this presentation, the dielectric properties of SiO_2 / BN composite is studied using the the logarithmic rule in the GEM equation.It is found that the dielectric constant of the composite material is effected by the contens of Si_2N_2O and temperature. And because the dielectric constant of SiO_2 is smaller and its variation is more stable, so when the content of BN is not high, the dielectric constant of SiO_2 / BN is lower and more stable. Also, it is found that the influence of frequency on the dielectric constant of the composite is relatively small, while its influence on the dielectric loss is more obvious, and when the temperature is relatively low, the influence is bigger, which is related to a bigger relaxation loss phase at a low temperature.

Introduction

The dielectric materials which have single ingredient more or less influence the performance, and in fact, in practical engineering application, they often mix dielectric materials which have different ingredients and get the materials which have a better performance, for example, the composite ceramic materials are a typical application[1]. Experiments show that the material system Si - B - O - N presents a better dielectric property, thermal stability and mechanical property than a single material (SiO_2, BN, etc.), and therefore, it is expected to become the wave-transmitting material which has the better performance at high temperature [2]. In this paper, the dielectric constant with temperature of SiO_2 / BN composite is studied by the logarithmic rule in the GEM equation, and the influence of components on the dielectric properties of composite is also analysized.

Procedures.

Calculating the dielectric properties of composite medium by theoretical modeling has an important practical significance. But because of the diversity of the structure of composite medium, it is very difficult to establish a calculation model which is correspondent with the actual situation of the composite medium, so we calculate the dielectric properties of composite medium mainly from the structure abstraction and dispersion particle regulation. The classic models of calculating the dielectric properties of composite medium are mainly Maxwell-Wagner Equation, generalized Bruggeman symmetric equivalent medium theory, generalized equivalent medium equation(GEM equation) and so on. Compared with Maxwell-Wagner Equation and generalized Bruggeman symmetric equivalent medium theory, GEM equation is more popular, and it forecasts the dielectric constant more accurate than the former two. And studies show that when there is a small difference of dielectric properties between the mediums, the factor of shape does not work. The dielectric properties of the materials of H - BN and SiO2 studied in this paper have a small difference, so when setting up the dielectric property model of the composite SiO_2 / BN, it should directly use the logarithmic rule in the GEM equation without considering the shape factor.

In the process of burning BN and SiO_2 into the ceramic, under certain conditions, there may generate some new phase. When the temperature reaches 1670K or so, BN reacts with SiO_2 and generates Si_2N_2O, namely[3]:

$$2BN + 2SiO_2 \xrightarrow{T \geq 1670K} Si_2N_2O + B_2O_3 \quad (1)$$

At the same time, with the increase of temperature, Si_2N_2O is also continuously decomposed, and under normal pressure, and at 1600 ° C, the decomposition reaction begins to become significant.

$$3Si_2N_2O(s) \longrightarrow 3SiO(g) + N_2(g) + Si_3N_4(s) \quad (2)$$

That is to say, when Si_2N_2O is generated, but at the same time, it is also decomposed, and its content is a process of dynamic change, so in order to simplify the analysis, we made the following assumptions:

(1) Si_2N_2O is generated from 1600K, and after that, its content gradually increases, until at 1900K, its content reaches 5% of the maximum. Afterwards, due to the intensive decomposition, its content will gradually decline, and it will be fully decomposed and generate SiO at 2100K. Because the content is small, we think that the content of SiN_2O presents a linear variation;

(2) SiO_2 participates in the reaction and generates Si_2N_2O, and we think that the content of SiO_2 in the system is invariable, namely, it only considers the mutual change of the content of BN and Si_2N_2O;

(3) Considering the change situation of the dielectric constant with temperature of three-phase system within the temperature scope from 1600K to 2100K, the dielectric constant of SiO_2, BN and Si_2N_2O respectively takes the dielectric content expression of quartz glass, solid BN and solid Si_2N_2O.

e_1, e_2, e_3 is the dielectric constant of SiO_2, BN and Si_2N_2O respectively, which the data are from our former studies[4-6]. And V_1, V_2, V_3 is their volume fraction respectively; the dielectric constant of composite is e_f .So, according to the logarithmic rule,

$$\lg e_f = V_1 \lg e_{SiO_2} + V_2 \lg e_{BN} + V_3 \lg e_{Si_2N_2O} \quad (3)$$

In this formula, the dielectric constant of three phases is taken according to the formula (3), $V_1 + V_2 + V_3 = 1$, as a parameter, V_1 can determined its numerical value by itself, and based on (1) and (2), the partial temperature interval volume fraction can be written as the following form:

When the temperature is at1600K-1900K,

$$\begin{cases} V_1 \\ V_2 = 1 - V_1 - V_3 \\ V_3 = \dfrac{5\%}{1900-1600}(T-1600) \end{cases} \quad (4)$$

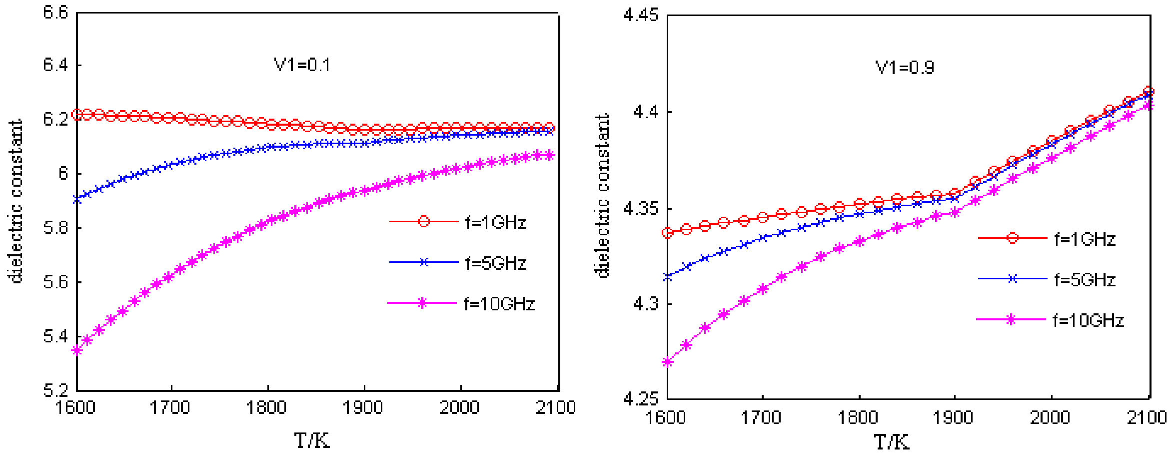
When the temperature is at1900K-2100K,

$$\begin{cases} V_1 \\ V_2 = 1 - V_1 - V_3 \\ V_3 = \dfrac{5\%}{2100-1900}(2100-T) \end{cases} \quad (5)$$

The generation of Si_2N_2O doesn't influence the content of impurities and hydroxyl, so we believe that the loss of compound system doesn't change because of the content of Si_2N_2O, and the logarithmic rule is also applicable for the loss, and therefore, we can get:

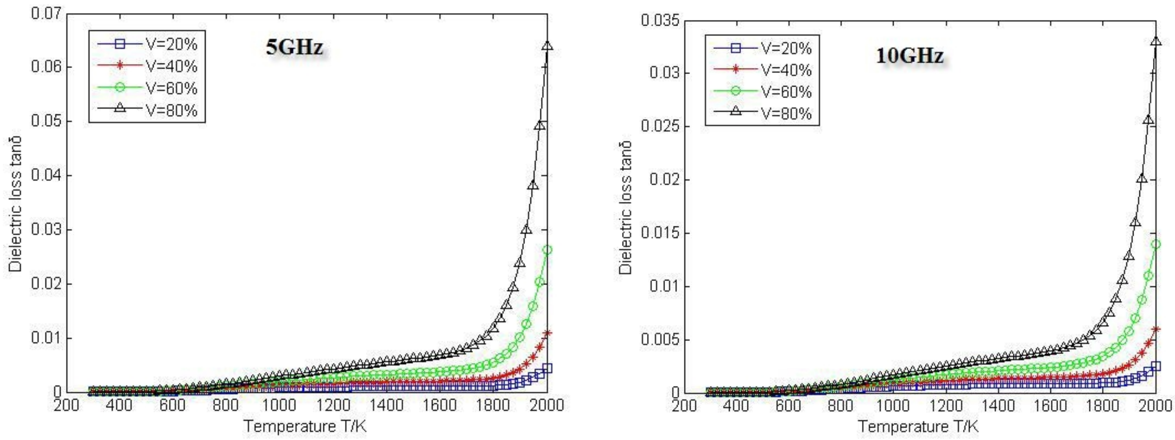
$$\log \tan \delta = V \log \tan \delta_{BN} + (1-V) \log \tan \delta_{SiO_2} \quad (6)$$

Results and Discussion. According to the above formulas, the dielectric properties of SiO_2 / BN composite can be obtained, shown in Fig.1 and Fig.2, which V_1 is the volume fraction of SiO_2. It is seen that Si_2N_2O has an influence on the dielectric constant of the composite, and at 1900K, we can see that there comes turning points in different degrees. When V_1=0.1, this variation is more prominent, and at the same time, we can also understand that this kind of variation can be weakened with the increase of frequency. At the same time, we can also see that the intermediate phase Si_2N_2O will be decomposed at a high temperature, and it has a smaller influence on the dielectric constant of complex medium, and when the frequency is high, this influence almost doesn't exist. Also, it can be seen from Fig.1-Fig.2 that, because the dielectric constant of SiO_2 is smaller and its variation is more stable, so when the content of BN is not high, the dielectric constant of SiO_2 / BN is lower and more stable. The frequency's influence on the dielectric constant of the composites is relatively small, while its influence on the dielectric loss is more obvious, and when the temperature is relatively low, the influence is bigger, which is related to a bigger relaxation loss phase at a low temperature.



(a) (b)

Figure 1 dielectric constant of SiO2 / BN composite (Fig.1 (a)V_1=0.1, Fig.1 (b)V_1=0.9)



(a) (b)

Figure 2 dielectric loss of SiO2 / BN composite (Fig.2(a) *f*=10GHz, Fig.2(b). *f*=10GHz)

Conclusions

When the temperature is between 300K to 1600K, the dielectric constant of compound system monotonically increases with the increase of temperature, but when the temperature is above 1600K to 1900K, with the generation of Si_2N_2O phase, the value of dielectric constant decreases, and when the temperature is above 1900K, the variation of the dielectric constant of the composite material decreases and the rate of descent is faster. On the one hand, with the decomposition of Si_2N_2O phase, the value of dielectric constant decreases; on the other hand, SiO_2 begins to melt, and the contribution of ion displacement polarization to the dielectric constant decreases, so that the dielectric constant decreases with the increase of temperature. Also, it can be seen that, because the dielectric constant of SiO_2 is smaller and its variation is more stable, so when the content of BN is not high, the dielectric constant of SiO_2 / BN is lower and more stable. The frequency's influence on the dielectric constant of the composite is relatively small. Similarly, the loss of the composite material is given priority to the relaxation loss at a low temperature, while at a high temperature, it is given priority to the electronic conductive loss, and in the middle temperature area, the impurity ion conductive loss plays a significant role.

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