Journal of Alloys and Compounds 509 (2011) 9279–9283

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jallcom

Influence of the various doping elements on the microwave dielectric loss of silica

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ARTICLE INFO

Article history:
Received 9 February 2011
Received in revised form 30 April 2011
Accepted 8 May 2011
Available online 24 June 2011

Keywords:
Dielectric loss
Impurities
Electrical conductivity
Activation energy

ABSTRACT

This article presents an evaluation of the influence of various doping elements on the dielectric loss of silica glass. Based on the Anderson–Stuart model, the temperature dependence of the electrical conductivity activation energy was investigated; then, comparisons of microwave dielectric loss of silica doped by various elements such as Li_2O, Na_2O, K_2O, CaO, MgO and Al_2O_3 are presented. At 1373 K, the dielectric loss of silica doped with Li_2O was found to be 10 times more than that the undoped one. The order in the impact of the doping elements on the microwave dielectric loss was found to be $Li_2O > Na_2O > K_2O > MgO > CaO > Al_2O_3$. This work is of importance with regard to applications that employ silica glass at conditions that involve especially high temperature and also in the microwave frequency range.

1. Introduction

Vitreous silica (v-SiO_2) has been one of the most extensively employed materials because of its potential application in ceramic and glass industries as well as in optical fibers, microelectronics and catalysis [1–3]. Material components and manufacturing technologies involved in the production of vitreous silica have been investigated with an aim to improve the practical performance of this material [4–7]. The spectroscopic and dielectric magnetic properties of the doped silica have been reported in the literature recently [8–10]. These authors believe that an understanding of how the electrical conductivity and the activation energy of a glass depend on the concentration of the species of electric charge carriers is very critical for glass scientists and technologists. Fortunately, the effects of alkali ions on the electrical conductivity of oxide glass have previously been investigated, with reports that the electrical conductivity is increased with increase in the concentration of alkali ions [11–14]. In former research works, glass samples were called silicate glasses, because the concentration of impurities was always more than 5%. However, there has been little information published on the exploitation of the influence of impurities present at low concentration (less than 1%) on dielectric properties, especially the microwave dielectric loss at high temperatures greater than 1000 K. Silica glass is known to inevitably contain impurities [15]. The nature and the concentration of impurities depend on the processes involved in its preparation. Due to the impurities' effects on various properties of the material such as its conductivity, when it is used under high temperature and high frequency, efforts should be made to purify the material to achieve better performance. If we only investigate several typical impurities' effects on the conductivity of the material, we can find certain impurity has a small impact on the conductivity of the material, so we do not have to take time and energy to eliminate it but only have to consider other impurities with bigger impacts. In this way, a large amount of time and money can be saved in practical application and it would be highly beneficial for the application of silica glass, especially at high temperatures (above 1000 K) and in the microwave frequency range.

It is well known that a model proposed by Anderson and Stuart (the A–S model) [16] is considered the most successful in explaining the meaning of physical parameters, such as ionic radii, relative dielectric permittivity and elastic modulus. Unfortunately, for the early A–S theory, there is little experimental data available to support it. Recently, Balaya et al. [17] successfully employed the A–S model to analyze a lead silicate system; similarly, Jogad [18] and Pan and Ghosh [19] applied the same model to analyze glass–ceramic and lead bismuthate glass systems, respectively.

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doi:10.1016/j.jallcom.2011.05.030

This study aimed to evaluate the influence of different doping materials on the dielectric loss of silica glass. Firstly, the temperature dependence of electrical conductivity activation energy of impurities was investigated in the Anderson–Stuart model [16]. Then, comparisons of the microwave dielectric loss of silica doped with various impurities, i.e., Li_2O, Na_2O, K_2O, CaO, MgO and Al_2O_3 are presented. Finally, available experimental data are applied to validate results obtained during calculation.

2. Methods

The disordered structure of glass provides a highly complex network for the transport of mobile ions. It is well known that the physical nature of silica glass is directly related to the integrity of networks, especially the amount of oxygen bridge bonds and non-bridging oxygen bonds. If the network structure is destroyed by metal oxides, it results in increase in the number of non-oxygen bridges and a reduction in the number of oxygen bridges; this, then, enhances the dielectric loss. In order to study the influence of the impurities on the dielectric loss of silica glass, certain related problems should be considered: (1) the status of various metal oxides, in general, in two types, i.e., the mobile ions or an atomic group and (2) the model can be used to express the impact of ionic radius and charge on the electrical conductivity and to study the effects of doping on the dielectric loss of silica.

For electromagnetic wave-absorbing materials, dielectric properties are represented by the relative complex permittivity, $\varepsilon' - j\varepsilon''$, where ε', the dielectric constant, describes the ability of a material to store energy and ε'', the dielectric loss factor, reflects the ability of a material to dissipate the energy of the electrical field. For silica glass, the total dielectric loss in the microwave band includes the relaxation and conduction losses. In general, the dielectric loss ($\tan\delta$) can be expressed by the following equation:

$$\tan\delta = \frac{\varepsilon''}{\varepsilon'} + \frac{\sigma_e}{\varepsilon_0 \varepsilon' \omega} + \frac{\sigma_{im}}{\varepsilon_0 \varepsilon' \omega} \qquad (1)$$

where σ_e is the electron conductivity, σ_{im} is the conductivity of impurity ions, ε_0 is the permittivity in vacuum and ω is the angular frequency. The first term is related to the relaxation loss whereas the second denotes conduction loss. The influence of impurities on the dielectric loss of silica glass mainly depends on the relaxation and ion-conduction losses. However, for glass systems, the ion-conduction loss plays a major role in influencing the dielectric loss, which tends to increase with the temperature, especially at high temperatures. It is well known that the ion-conduction loss is mostly affected by the ion conductivity, which is given by

$$\sigma_{im} = \frac{n_0 q^2 l^2 \nu}{6kT} e^{-W_S/kT} \qquad (2)$$

where n_0 is the number densities of the impurities and l is the jumping distance of the impurity ions. The W_S is the activation energy of conductivity of impurities, and is discussed in Section 3.1. The ionic conductivity of pure silica is not particularly high because of the presence of few impurities. At sufficiently high concentrations, most impurities can change the structure of glass; in this less compact form, the conductivity of mobile ions is not determined by only their activation energy. In this study, the authors focus on the dielectric properties of silica doped with low concentrations of impurities.

Therefore, by drawing from Eqs. (1) and (2), the ion-conduction loss can be expressed as:

$$\tan\delta_{rim} = \frac{\sigma_{im}}{\omega \varepsilon_0 \varepsilon'} = \frac{n_0 q^2 l^2 \nu}{6kT\omega \varepsilon_0 \varepsilon'} e^{-W_S/kT} \qquad (3)$$

3. Results and discussion

3.1. The influence of temperature on activation energy

If impurities comprise alkali metal oxides or alkali earth metal oxides, the network structure is modified by the presence of interstitial positive ions and the incorporation of corresponding oxygen ions; this results in the disintegration of the three-dimensional network structure of Si–O tetrahedrons, i.e., the network modifier (N.W.M.). An electrical field may increase the probability that positively charged ions would move along the direction of the field by an effective alteration in the height of a potential barrier. The resulting net migration of the positive ions accounts for the flow of electrical charge through the silica glass. Therefore, the impact of alkali metal ions on the dielectric properties of silica glass mainly depends on the increase in ionic conductivity. From Eq. (2), it can be observed that the activation energy W_S should be known in advance in order to obtain the ionic conductivity.

Table 1
The activation energy of impurities of silica.

Ion	Li^+		Na^+		K^+		Mg^{2+}		Ca^{2+}	
	E_b	E_S	E_b	E_S	E_b	E_S	E_b	E_S	E_b	E_S
300 K	1.0	0.08	0.79	0.43	1.62	0.44	0.87	1.72	1.53	0.45
1400 K	0.98	0.09	0.75	0.45	1.59	0.7	0.84	1.84	1.48	0.48

Based on the Anderson–Stuart model, the activation energy for conduction was considered the energy E_b to overcome electrostatic forces and the energy E_S to open up "doorways" in the structure that were sufficiently large for ions to pass through. Thus, E_b represents the energy required to remove a cation from a non-bridging oxygen site, and E_S describes the expansion of the structure as the ion moves from one site to another. In addition, the activation energy is given by the equation:

$$W_S = E_b + E_S = \frac{\beta z z_0 e^2}{\gamma(r + r_o)} + 4\pi G r_D (r - r_D)^2 \quad \beta = \frac{2.1 - r}{3.5} \qquad (4)$$

where z, r are the charge and radius of ions, z_0, r_0 are the charge and radius of oxygen ions, r_D is the effective radius of the doorway, G is the shear modulus of doped silica glass and γ accounts for the deformability of the oxygen atom. A covalent bond implies the presence of electron-cloud deformation and, therefore, γ is called the covalence parameter, and would be equal to the dielectric constant of the glass [16].

From Eq. (4), it is also evident that the key parameters are the elastic modulus (G), a 'door way' (r_D) and the relative dielectric permittivity (γ), which indicates the degree of charge neutralization between the ion and its immediate neighbors. The mobility of impurity ions is altered with increase in temperature and concentration of impurities; thus, G and r_D represent functions of temperature and concentrations of impurities. The relation between the shear modulus and temperature from room temperature to approximately 1573 K can be determined from a group of vitreous silica ($dG/dT = 18.5 \times 10^{-5}$/K) [20]. In addition, the shear modulus G_0 (the value of the shear modulus at 300 K) [21–24] showed a small reduction with increase in sodium composition. However in this work, it is presumed that there is no significant influence by very small quantities of impurities on the shear modulus of silica glass; thus, G_0, in this instance, is 3.1 GPa. The researcher Nascimento [25] reported that r_{D0} (at 300 K) versus composition varied in a range, i.e., $0.37\,Å < r_{D0} < 0.72\,Å$. Therefore, for small amounts of doping, it can be deduced that the value of r_{D0} is approximately 0.37 Å. By considering the possible expansion of doorway as a result of temperature, we obtain:

$$r_D(T) = [1 + \alpha_1 (T - T_0)] r_{D0} \qquad (5)$$

where α_1 is the thermal coefficient of silica glass ($\alpha_1 = 5 \times 10^{-7}$) [26]. The relative dielectric permittivity γ was found to increase monotonically with increase in sodium concentration and the temperature [27–29], and thus:

$$\gamma = \gamma_0 + \frac{d\gamma}{dT} T + \frac{d\gamma}{dn} n \qquad (6)$$

where γ_0 is 3.82, $d\gamma/dn$ is 0.1681 ± 0.0029/mole% [25] and $d\gamma/dT = 3.6 \times 10^{-4}$/K [26]. Thus, the temperature dependence of the activation energy is expressed as:

$$W_S = E_b + E_S = \frac{\beta z z_0 e^2}{[\gamma(r + r_0)]} + 4\pi G_0 \left[1 - \frac{dG}{dT}(T - T_0)\right] r_D(T) \times [r - r_D(T)]^2 \qquad (7)$$

From Eq. (7), the binding energy and strain energy of the impurities at different temperatures are estimated and the results are

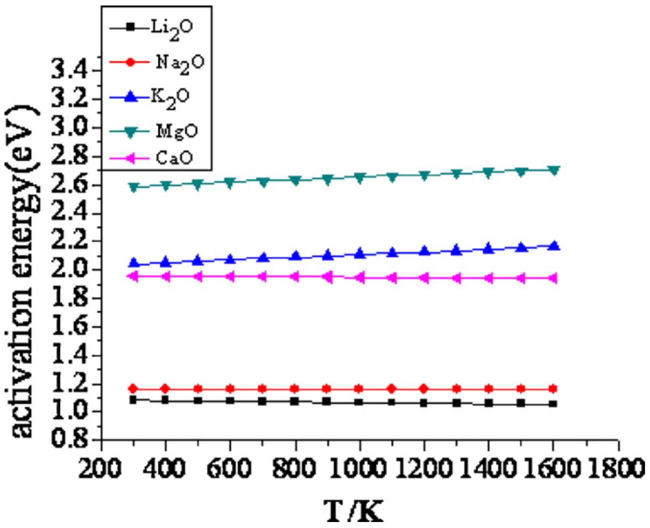

Fig. 1. The temperature dependence of activation energy of impurities. The calculated optical properties for Si_2N_2O.

listed in Table 1. The activation energy of alkali metal ions are evidently lower than that of alkali earth metal ions; this is because the activation energy is mainly determined by the binding energy, which is definitely affected by the valence of impurities. Further, the binding energy is found to be increased with the ionic radius because the smaller radius of the ions facilitates closer association with oxygen. Moreover, It should be noted that the strain energy decreases in the order from Li^+ to Na^+ and then K^+; this is attributed to the increase of ionic radius [Pauling's ionic radii of alkali metal ions is Li^+(0.60 Å), Na^+(0.95 Å) and K^+(1.33 Å), respectively], which makes transportation difficult and results in lower contribution to electrical conductivity.

The dependence of activation energy with different temperatures for alkali metal ions and alkali earth metal ions is shown in Fig. 1. The activation energy of Li_2O, Na_2O, and CaO decreases with temperature. However, the activation energy with other ions increases with the temperature, and this is ascribed to the different temperature coefficients of binding energy and strain energy. The permittivity (γ) increases with increasing temperature. Therefore, it can be noted from Eq. (4) that the binding energy decreases as a result of the above properties. However, the enhancement of strain energy will induce an increase in the shear modulus with the temperature. Therefore, if the radius of impurity ions is smaller, such as in the case of Li_2O, Na_2O and CaO, the binding energy would be greater than the strain energy; this results in the decrease of activation energy with temperature because the increase of the strain energy cannot compensate for the decrease of the binding energy. Similarly, when the radius of impurities is greater, an opposite trend becomes evident.

However, the substitution of Al_2O_3 for v-SiO_2 results in more complex problems. The coordination number of the Al^{3+} ion varies between 4 and 6. The findings for [AlO_4] coordination are similar to that of the [SiO_4] coordination. An Al^{3+} ion can replace a Si^{4+} ion in the glass structure. The necessary valency compensation can be accomplished by externally introduced alkali ions; this leads to the formation of bridging oxygen. In instances with the [AlO_4] replacement, spaces of separation are become closed throughout the glass structure, which is again strengthened [30]. However, for small concentrations of Al_2O_3 (<0.5%), Yoldas [31] reported that Al^{3+} ions exist within [AlO_6] groups in glass networks. Further, the Al_2O_3 is called an intermediate compound because is incorporated into the networks without deforming glass.

Therefore, the impurities evaluated in this study are classified as: (a) mobile ions, such as alkali metal oxides (Li_2O, Na_2O, K_2O, etc.) and alkali earth metal oxides (CaO, MgO, etc.) and (b) intermediate compounds such as Al_2O_3.

3.2. *The effects of impurities on the dielectric loss of silica*

The impact of impurities on silica glass mainly includes relaxation and ion-conduction losses at microwave frequencies. From Eq. (1), it is evident that the activation energy for conduction is more or less equal to that required for the relaxation time. This is because an ion has to overcome the same barrier while conducting as well as relaxing; this finding is consistent with the fluctuation–dissipation theory. Thus, the influence on dielectric loss of silica doped with 50 ppm R_2O (R = Li, Na or K) and RO (R = Ca or Mg) up to a temperature of 1600 K, at frequency of 10 GHz is calculated by using Eqs. (1)–(7); the results are shown in Fig. 2. At 1373 K, the dielectric loss doped with Li_2O is found to be 10 times more than that the undoped silica. The losses of these impurities, with the exception of Li_2O, are given in the inset in Fig. 2 for comparison. From Fig. 2, we observe that R_2O has a greater influence on the dielectric loss of silica than RO at the same concentration. This is because R_2O is bound less strongly to the surrounding oxygen atoms. Therefore, an increase in the number of migrating ions can, most probably, be expected as temperature increases. However, it also shows that doping effects on dielectric loss are not significant when the temperature is less than 800 K due to activation of fewer ions. Moreover, it is identified that the dielectric loss becomes large with increase in the concentration of impurities and temperature. In this scenario, the change in activation energy can be attributed to two aspects. On one hand, in the absence of N.W.M. ions, the structure is a continuous network comprised of Si^{4+} and O^{2-} ions. The O^{2-} ions are easily deformed. Therefore, glassy networks may be considered a collection of positive charges and highly deformed negatively charged clouds that represent the O^{2-} ions. The bonding of this arrangement is constituted of ionic and covalent bonds in varying measure. To the extent that the network is not influenced by N.W.M. ions, the O^{2-} electron clouds continue to take up symmetrically distorted configurations. When an N.W.M. ion, or an extra charge, is introduced to the networks, the neighboring oxygen electron clouds become highly distorted. From this, it can be inferred that the bond between the N.W.M. ion and oxygen atoms must be partially covalent. In the discussion earlier, γ was called

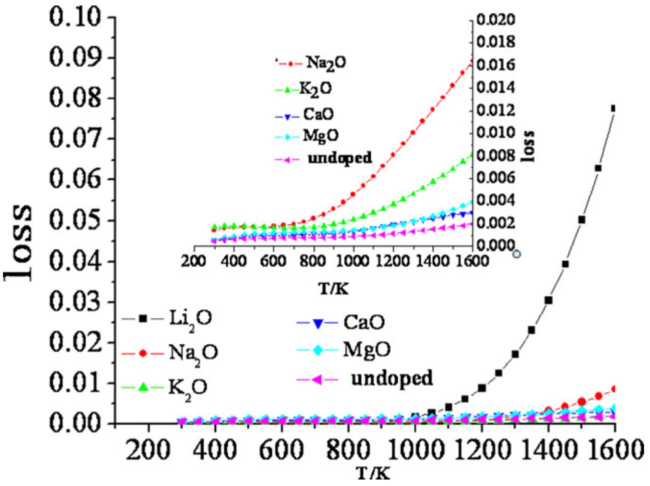

Fig. 2. The dieletric loss of silica with RO and R_2O. The inset shows the loss of RO and R_2O except Li_2O.

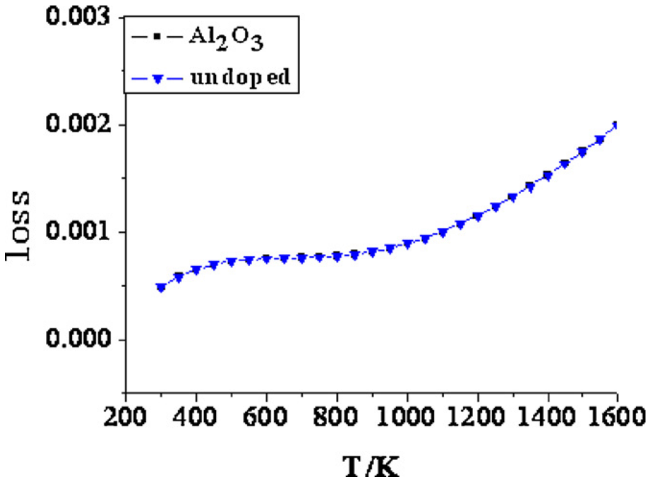

Fig. 3. The dielectric loss of silica doped Al_2O_3 of 200 ppm.

the covalence parameter, and it accounted for the deformability of the oxygen atoms. When sufficient N.W.M. ions are doped or the temperature increases to a sufficient level, the deformability of the oxygen atom also increases. This results in a higher value of γ. Consequently, the binding energies are expected to become reduced. On the other hand, the strain energy may reduce with decrease in temperature and concentrations of impurities due to the lower height of a potential barrier.

Specifically, for the Al_2O_3 dopant at low concentrations, as indicated from the above discussion, there exist $[AlO_6]$ groups in networks of silica glass because of the close association between Al and O. In such a case, the dopant can be treated as a compound, and the overall dielectric loss can be evaluated by the following equation:

$$\tan\delta = V_l \tan\delta_l + V_h \tan\delta_h \qquad (8)$$

where the subscript h and l represent Al_2O_3 and SiO_2, respectively. Here, V is the content and $\tan\delta$ is the dielectric loss. Therefore, using Eq. (8), the dielectric loss of silica doped with Al_2O_3 can be calculated, as shown in Fig. 3. However, when compared with relatively large losses of the R_2O-doped silica glass, the loss in the Al-doped silica is quite small.

Fig. 4(a) summarizes the influence of two types of impurity oxides on the doped silica glass. From Fig. 4(a), it can be observed that impurity oxides such as R_2O are regarded as the “fast” ions because they may result in “open” structures of the networks as well as increased electrical conductivity. Further, Al_2O_3 is defined as one of the “forbidden” ions because it may result in the formation of “closed” structures within the network. For alkali-containing glass, it is observed that small amounts of Al_2O_3 may induce an elevation of electrical resistance [31]. Further, Fig. 4(b) shows the change in the jumping distance when Al_2O_3 is introduced into the alkali-containing glass. The Al^{3+} can be observed to reduce the jumping distance of the alkali ions because of the existence an atomic group in the network. From Eq. (3), it can be observed that the jumping distance of the impurity ion (l) can affect the dielectric loss significantly. In addition, the introduction of Al_2O_3 may hinder the mobility of alkali ions, and then decrease the dielectric loss of the glass.

3.3. Comparison between calculations and experimental data

A silica glass sample with a density of 2.2 g/cm^3, doped with 2000 ppm of Al_2O_3, was selected to measure its permittivity within a microwave frequency (10 GHz) by the cavity-perturbation technique at temperatures ranging from 300 K to 1573 K. The details of this technique have been reported in the reference [32]. Results of comparison between calculated and measured parameters are presented in Fig. 5. It is evident that results of calculation are close to results obtained from experimental data; further, it is evident that Al_2O_3 has little impact on the dielectric loss, and this finding validates the assumption that it can be considered as a compound equivalent to those discussed in the above analyses. The dielectric loss was observed to decrease with temperature from 300 K to 1000 K; this can, doubtless, be ascribed to the absorbed moisture. At room temperature, the dielectric constant and loss from absorbed water are large. With increase in temperature, the absorbed water volatilizes rapidly; this results in the decrease of loss. Under the same doping concentration and temperature conditions, the influence of impurities on the dielectric loss of silica glass can be placed in the order $Li_2O > Na_2O > K_2O > MgO > CaO > Al_2O_3$. It is noteworthy that, from phenomenological analyses undertaken in the present study, Li^+ is an important conductivity-enhancing composition, and its doping concentration in silica systems should be strictly controlled in order to obtain a lower dielectric loss at high temperatures in practical applications.

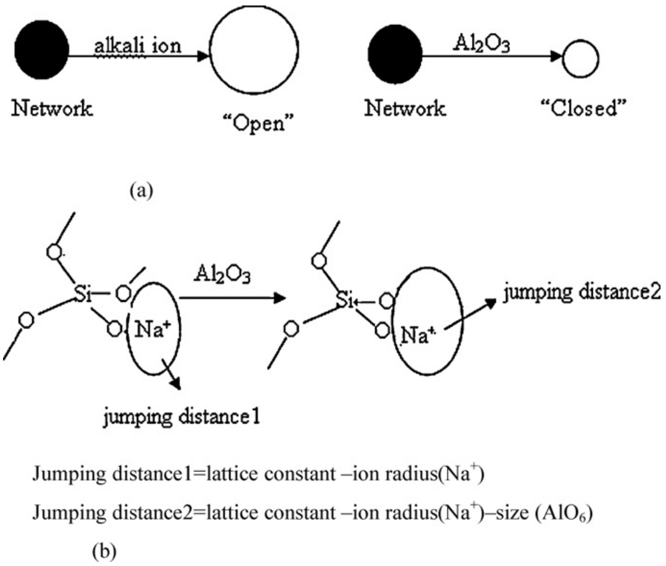

Fig. 4. (a) A schematic representation of the influence of two impurity oxides on the glass. (b) The representation of the addition of Al_2O_3 in alkali-containing glass.

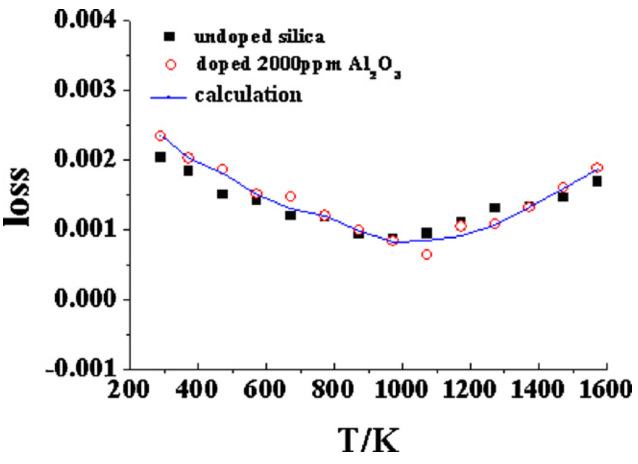

Fig. 5. The dieletric loss of silica with 2000 ppm Al_2O_3.

4. Conclusions

In summary, the results obtained in this study can be summarized as:

(a) The activation energy of Li_2O, Na_2O and CaO decreases with temperature; however, K_2O and MgO showed an opposite trend which was attributed to different temperature coefficients of binding energy and strain energy. From comparisons of the microwave dielectric loss of silica doped by various impurities, the influence of impurities on the microwave loss can be defined in the order: $Li_2O > Na_2O > K_2O > MgO > CaO > Al_2O_3$.
(b) Phenomenological analyses reveal that Li^+ is an important conductivity-enhancing constituent in silica systems; therefore, its doping concentration should be strictly controlled in order to obtain a lower dielectric loss at high temperatures, in practical applications.
(c) The existing experimental data validate results of calculation, and indicate that Al_2O_3 has little impact on the dielectric loss.

Acknowledgements

We gratefully acknowledge the support of the National Science Foundation of China under Grant No. 50902017, Scientific Reserch Fund of SiChuan Provincial Education Department (No.09ZC029), Chengdu Medical Foundation (No.CYZ08-016).

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