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Microwave properties of vanadium borate glasses

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Abstract. **A.c. conductivity, dielectric constant and loss, and variation with temperature (302–373 K) for four different compositions of V_2O_5–B_2O_3 glasses were reported at 9·586 GHz microwave frequency. The quality factor (*Q*) and attenuation factor (*a*) being the important parameters in the microwave range of applications were also studied. The change in the dielectric constant and loss was observed with composition of V_2O_5. The maximum loss was found to be at 15V_2O_5 mol%. The peak was observed in loss with temperature.**

Keywords. **Semiconducting glass; V_2O_5–B_2O_3; microwave properties.**

1. Introduction

Ceramics and glasses possessing high relative dielectric constant, e' (~ 20 ~ 90), high quality factor, Q (> 3000–35000 at 3 GHz), and very small temperature coefficient of frequency (TCF) (~ 2 ppm/°C) in the microwave region are useful for exploiting the dielectric resonators in microwave applications. Ceramics and glass dielectrics might cheaply produce the microwave filters and oscillator components. In order to reduce the size of the components used in cellular telephones and other communication systems the multilayer structures have been developed today (Hirade 1992). The important factor is the lowering of sintering temperature of the dielectric ceramics. Choosing the low melting glass addition in dielectric ceramic, chemical processing and the smaller partial sizes can do the reduction in the sintering temperature. The starting materials B_2O_3, SiO_2, ZnO–B_2O_3 glass and other glasses can be used as glass additions in the microwave dielectric ceramics such as (Zr, Sn) TiO_4 etc.

The resonant cavity and resonant post frequencies have been evaluated and limiting conditions are determined in high silica microporous glass at 5·5–11·4 GHz (Yamamoto *et al* 1989). The dielectric constant and loss at these microwave frequencies have been reported.

The effect of glass former additions on BaO–TiO_2–WO_3 and (Zr, Sn) TiO_4 microwave ceramics was studied by Takada *et al* (1994). The chemical reaction between ceramics and glass phases responsible for sudden drop in the quality factor of microwave dielectric ceramic at sintering temperature near 1000°C was discussed. Nivias and Green (1976) studied dielectric properties of aluminoborosilicate glasses at microwave frequencies. These glasses have higher glass transition temperature, T_g. Very few reports are available on glasses in microwave region. The RF and microwave conductivity of bulk chalcogenides were studied by Cleach and Palmier (1979). The results are analysed in terms of Pollak and Pike (1972) two-level tunneling model.

Keeping in view of these applications of glasses in the microwave ceramics it is imperative to investigate various glasses in the microwave frequency region. The work in this regard is found to be scanty. Therefore, it was decided to study the V_2O_5–B_2O_3 glass system at the microwave frequency of 9·586 GHz in X-band with an intention to get new insight into the dielectric properties as well as the structural defects.

2. Experimental

2.1 *Preparation of glasses*

Semiconducting glasses, V_2O_5–B_2O_3, of varying thickness of rectangular shape 0·9 × 0·4″, which is the size of rectangular waveguide in X-band microwave bench, were prepared in the laboratory by sudden quenching technique (Yawale and Pakade 1993). Powdered form of (Analar-grade) boric oxide and vanadium pentoxide were taken in proper proportion. Weighing was done on monopan (K-ROY) balance having an accuracy of ± 10 μg. These two powders were finely mixed for 15 min using mortar and pestle, to form a homogeneous mixture. This mixture was transferred to fire clay crucible which was then subjected to melting at temperature 900°C ± 10°C in an automatic temperature controlled muffle furnace. The duration of melting was generally

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150 min. The homogeneous molten mass was quickly cast into mould of stainless steel (0·9″ × 0·4″ for X-band) at room temperature. The glass samples were then formed by quenching the melt.

Using the above method, four types of samples of composition ($B_2O_3 : V_2O_5$) 95 : 5, 90 : 10, 85 : 15 and 80 : 20 were prepared. For each type of glass, 3–4 samples of varying thickness were prepared. The general formula is

$$XB_2O_3 - (100 - X)\ V_2O_5.$$

The glasses of composition > 20% V_2O_5 never formed. Increasing the concentration beyond 20 mol% increases the cavity in the samples which makes them very brittle. The glass surface of all the samples was made plane by polishing with fine emery paper for perfect reflection of electromagnetic wave. Thickness of samples was measured by using screw gauge having an accuracy ± 0·001 cm, it was in the range of 0·3–1·2 cm. The amorphous nature of the sample was checked by X-ray diffraction spectra.

2.2 *Dielectric constant measurement at microwave frequency*

For accurate measurement of dielectric constant width at twice minima method was used (Lance 1964). Following steps were followed in this method.

(i) The frequency of source was determined by taking micrometer reading of cavity resonator.
(ii) The guide wavelength, l_g, was measured by short circuiting waveguide section.
(iii) The distance, ΔX, shown in figure 1 was measured by width at twice minima method.
(iv) The position of minima without sample ‘A’ was recorded (figure 1).
(v) Dielectric sample (glass) was then introduced in the waveguide touching the short-circuit end.
(vi) The distance, ΔX_s, was measured.
(vii) The position of minimum ‘B’ of standing wave pattern occurring with sample was recorded (figure 1).
(viii) Shift in minima Δl was calculated.

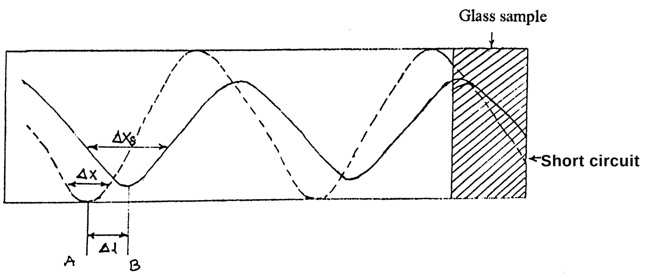

Figure 1. Standing waves in the waveguide with and without sample.

The dielectric constant was calculated by using the formula given in (1). The readings were noted in the temperature range 302–373 K.

3. Theory

3.1 *Dielectric constant*

Dielectric constant (e') is given by the following formula

$$e' = (X l_0/2pd)^2 + (l_0/l_c)^2, \tag{1}$$

where, l_0 is the free space wavelength (= 3·03 cm), l_c the cutoff wavelength (= 4·582 cm), d the thickness of the sample and X the multivalued function and its value was chosen from the tables of ($\tan X/X$) given by Von Hippel (1954).

3.2 *Loss tangent* (*tan d*)

The loss tangent was calculated using the formula (Lance 1964)

$$\tan d = \{|\Delta X_s - \Delta X|/e'd\}\ (l_0/l_g)^2. \tag{2}$$

3.3 *Loss factor* (e^2)

The loss factor, e'', is determined from the following formula

$$e'' = e' \tan d. \tag{3}$$

3.4 *Microwave conductivity* (s)

The microwave conductivity, s, of glass sample was determined from the following formula

$$s = (fe' \tan d)/1{\cdot}8 \times 10^{12}, \tag{4}$$

where, f is the microwave frequency (9·586 × 10^9 Hz).

3.5 *Attenuation* (a)

The attenuation (a) is given by

$$8{\cdot}686\ a = 8{\cdot}686\ (2p/l_0)\ [(1/2)\ e' \{\sqrt{1+\tan^2 d} - 1\}]^{1/2}\ (\mathrm{d}b/m). \tag{5}$$

4. Results and discussion

The dielectric constant, loss factor, microwave conductivity and attenuation are studied at microwave frequency (9·586 GHz) at different temperatures for glasses of V_2O_5–B_2O_3 in the range of composition 5–20 mol% of V_2O_5. The values of dielectric constant (e') and dielectric loss (e'') are found to be reasonable to the glasses (Von-Hippel 1954).

Table 1 reports the values of tan x/x for different thicknesses of the various glasses at room temperature, 302 K. The values of x are determined from tan x/x values and the average value of x is calculated to find out the dielectric constant, e'. Such values are also determined for other temperatures. It is observed that the values of x are near about same for different thicknesses of glass of same composition.

Figure 2 shows the variation of dielectric constant (e') with temperature for different compositions of V_2O_5. The value of dielectric constant varies between 12·4 and 14·7 for all the glass samples in the specified temperature range. It is observed that the variation of dielectric constant with temperature is small. Initially the value of e' is high at room temperature and then decreases and remains constant for further increase in temperature, but the dip is observed between 320 and 340 K. The behaviour of dielectric constant in all the compositions of glasses is similar. This type of behaviour is in quite agreement with the reported behaviour (Von-Hippel 1954) in the other glasses.

Table 1. Values of tan x/x for different thicknesses of the various glasses at 302 K.

Sl. no.	Sample V_2O_5–B_2O_3 (mol%)	Thickness (cm)	tan x/x	x
1	05–95	0·431	0·04345	3·295
		0·440	0·04746	3·300
		0·472	0·06440	3·360
2	10–90	0·417	0·0467	3·295
		0·427	0·0660	3·360
		0·447	0·0505	3·305
3	15–85	0·398	0·0533	3·315
		0·408	0·0521	3·321
		0·420	0·0490	3·300
		0·425	0·0532	3·315
		0·467	0·0575	3·330
4	20–80	0·360	0·0485	3·300
		0·406	0·0528	3·315
		0·437	0·0492	3·305
		0·470	0·0606	3·340
		0·553	0·0516	3·310

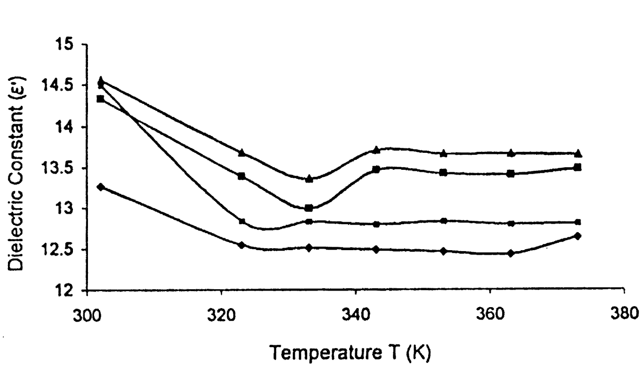

Figure 2. Variation of dielectric constant (e') with temperature (T) for different compositions of V_2O_5: (♦), $5V_2O_5$; (■), $10V_2O_5$; (▲), $15V_2O_5$ and (▮), $20V_2O_5$.

The variation of dielectric constant with composition of V_2O_5 is shown in figure 3 at room temperature and 373 K. The dielectric constant is found to be maximum at $15V_2O_5$ glass and decreases with increase in temperature.

Figure 4 shows the variation of dielectric loss (e'') with temperature for different compositions of V_2O_5. The zigzag nature is observed in the curve and for each composition the peak value of e'' is observed at a particular temperature. The loss decreases with increase in temperature after peak. The peak is observed at 335, 343, 353 and 373 K for 5, 10, 15 and 20 mol% of V_2O_5 glasses, respectively. The peaks are due to dipole mechanisms and may be explained on the basis of dipole losses.

The dielectric loss (e'') observed is maximum in $15V_2O_5$ at room temperature (302 K). This variation is shown in figure 5. At a temperature of 373 K, no peak is observed hence the maximum losses are in $15V_2O_5$–$85B_2O_3$ glass at microwave frequency. After 373 K, the loss decreases with increasing composition of V_2O_5. This

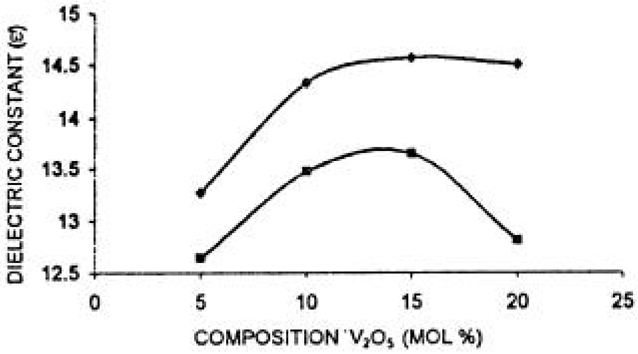

Figure 3. Variation of dielectric constant (e') with composition of V_2O_5 at room temperature (302 K(♦) and 373 K (■)).

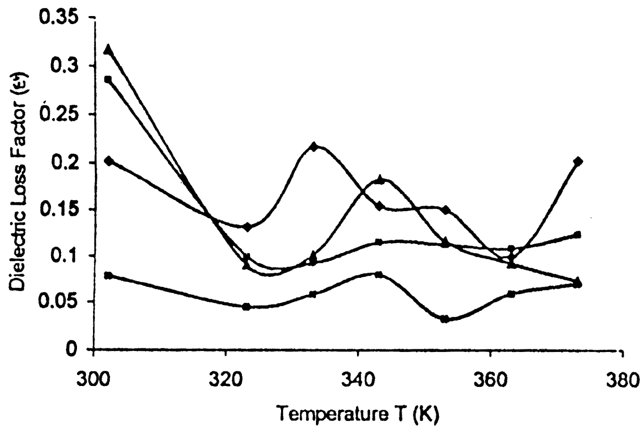

Figure 4. Variation of dielectric loss (e'') with temperature (T) for different compositions of V_2O_5: (♦), $5V_2O_5$; (■), $10V_2O_5$; (▲), $15V_2O_5$ and (▮), $20V_2O_5$.

Table 2. Values of microwave conductivity (s), quality factor (Q) and attenuation (a) at various compositions and at room temperature (302 K).

Sl. no.	Glass composition V_2O_5–B_2O_3 (mol%)	Microwave conductivity (s) $(\text{ohm-cm})^{-1}$ 10^{-4}	Quality factor (Q)	Attenuation (a) (db/cm) 10^{-2}
1	05–95	10·75	65·70	5·745
2	10–90	15·17	50·27	7·802
3	15–85	16·83	46·04	8·537
4	20–80	4·14	185·90	2·122

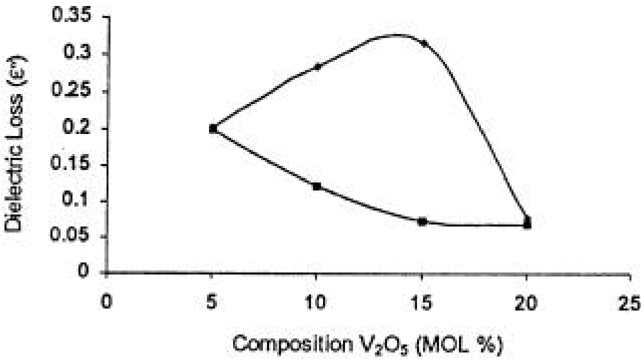

Figure 5. Variation of dielectric loss (e'') with composition of V_2O_5 at room temperature (302 K(♦) and 373 K (■)).

is the typical behaviour that has been observed in the V_2O_5–B_2O_3 glasses.

Table 2 reports the microwave conductivity, quality factor and attenuation at different compositions of V_2O_5 at room temperature (302 K). These values are found to be of the order of glasses in general.

The microwave conductivity (s) is evaluated from the dielectric loss (e''), therefore, the conductivity behaviour is observed to be same as in dielectric loss. At room temperature, $15V_2O_5$ glass shows maximum conductivity.

Generally the a.c. conductivity of the glasses may be explained on the basis of electron hopping among localized states. In lower frequencies, the measured a.c. conductivity has no true value due to electrode polarization and other problems. While measuring the microwave a.c. conductivity these problems are totally avoided due to electrode-less measurement. The Pollak-Pike (1972) model gives satisfactory results. By considering $f \sim 9$ GHz, $t_0 = 10^{-3}$ s and the value of conductivity as 10^{-4} $(\text{ohm-cm})^{-1}$, the density of states is found to be of the order of 10^{19} cm^{-3} which is quite reasonable.

The attenuation (a) at microwave frequency is measured at room temperature (302 K). The attenuation (a) increases with compositions of V_2O_5 and becomes maximum at $15V_2O_5$ and then decreases.

The microwave technique is electrode-less technique of measurement. Therefore, the measurements at microwave frequencies are free from electrode polarization and associated problems and thus give real material property. A good microwave dielectric should possess high relative dielectric constant ($e' \sim 90$), high quality factor, Q (> 3000–35000 at 3 GHz) and very small temperature coefficient of frequency (TCF). Having such properties at microwave frequency to the ceramic materials, then the material can be considered as a good microwave dielectric. To obtain these properties, it becomes necessary to reduce the sintering temperature of the dielectric ceramics. For this purpose the low melting glass addition including B_2O_3, SiO_2, ZnO–B_2O_3 glass etc plays an important role. The dielectric constant found here is < 20 and quality factor, Q is < 3000 which suggests that the glass, V_2O_5–B_2O_3 will not be useful as a good microwave dielectric material. But this is useful in other applications such as in semiconducting components.

5. Conclusions

The microwave properties of V_2O_5–B_2O_3 glasses have been investigated at 9 GHz and various temperatures. The dielectric constant, loss, quality factor etc are found to be composition dependent of V_2O_5. Not much variation in the dielectric constant is observed with temperature. The dielectric constant and quality factor values are not observed within the range of microwave dielectric. Hence, vanadate system is not useful as dielectric resonators in the microwave applications. In general at microwave frequency the movement of network and modifying ions are held responsible for relative dielectric constant and the oscillations between them for losses.

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