Compositional Effects on Thermal and Electrical Properties of Dielectric Glass Paste

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Abstract

The effect of Al_2O_3 on the dielectric constant and the coefficient of thermal expansion of lead borosilicate glasses for the application of PDP glass paste was investigated. Measurements were made theoretically by using empirical equations on the composition of the of glasses in which the PbO/Al_2O_3, B_2O_3/Al_2O_3 and SiO_2/Al_2O_3 ratios were systematically varied. As a result, with increasing PbO/Al_2O_3 the thermal expansion coefficient and the dielectric constant noticeably increased, while the change of B_2O_3/Al_2O_3 and SiO_2/Al_2O_3 ratios did not affect those properties of the glasses.

Introduction

Flat display panel is becoming a major new application for high-performance glass because this device offers advantage over CRT in terms of less weight and smaller size. The PbO-SiO_2-Al_2O_3-RO (R_2O_3) glasses have been the most commonly used system for frit glass and glass paste in plasma display panel (PDP) industry [1,2]. The main different composition of glass paste for double layers (upper and lower) is generally the existence of Al_2O_3: upper layer does not contain Al_2O_3, but lower one does. In the glass paste, Al_2O_3 can affect the electrical and the thermal properties of dielectric and act as an intimidator for the substrate glass, which is a soda-lime-silica glass with a softening temperature of 723°C [3]. The reasonable dielectric for PDP should have the same thermal expansion coefficient with the substrate and high dielectric constant as well as high thermal conductivity. For the development of an ideal dielectric for PDP, it would require several properties: to prevent formation of air bubbles, to make transparency above 80% after firing, to increase voltage with a thin film, and to match very well with bus electrode. To develop a new dielectric glass paste for low temperature firing, however, design of glass composition is the most important stage for the purpose.

In this work, we limited the measurements, dielectric constant and thermal expansion coefficient in the various properties. The optimum conditions, in producing dielectric layer for PDP, which have high dielectric constant and low thermal expansion coefficient are theoretically investigated only as a function of compositions, especially different oxide ratios PbO/Al_2O_3, B_2O_3/Al_2O_3 and SiO_2/Al_2O_3.

Theoretical background

It can be easily obtained on the optimum conditions of electrical and thermal properties based on a mixture rule that each property is closely related with their composition [4].

- Dielectric constant

$$\varepsilon = 10^{-2}\Sigma\, \varepsilon_i\, (\text{mol\%})_i \qquad (1)$$

- Coefficient of thermal expansion

$$\alpha = \Sigma\alpha_i\, (\text{wt\%})_i \qquad (2)$$

where ε_i and α_i are the additivity factors or the property coefficients for a given component i. The compositions considered in this work were varied in the following range: PbO (60-70wt%), B_2O_3 (10-25wt%), SiO_2 (5-20wt%) and Al_2O_3 (5-25wt%). Compositions of PbO-B_2O_3-SiO_2-Al_2O_3 glasses with variable ratios are in Table 1.

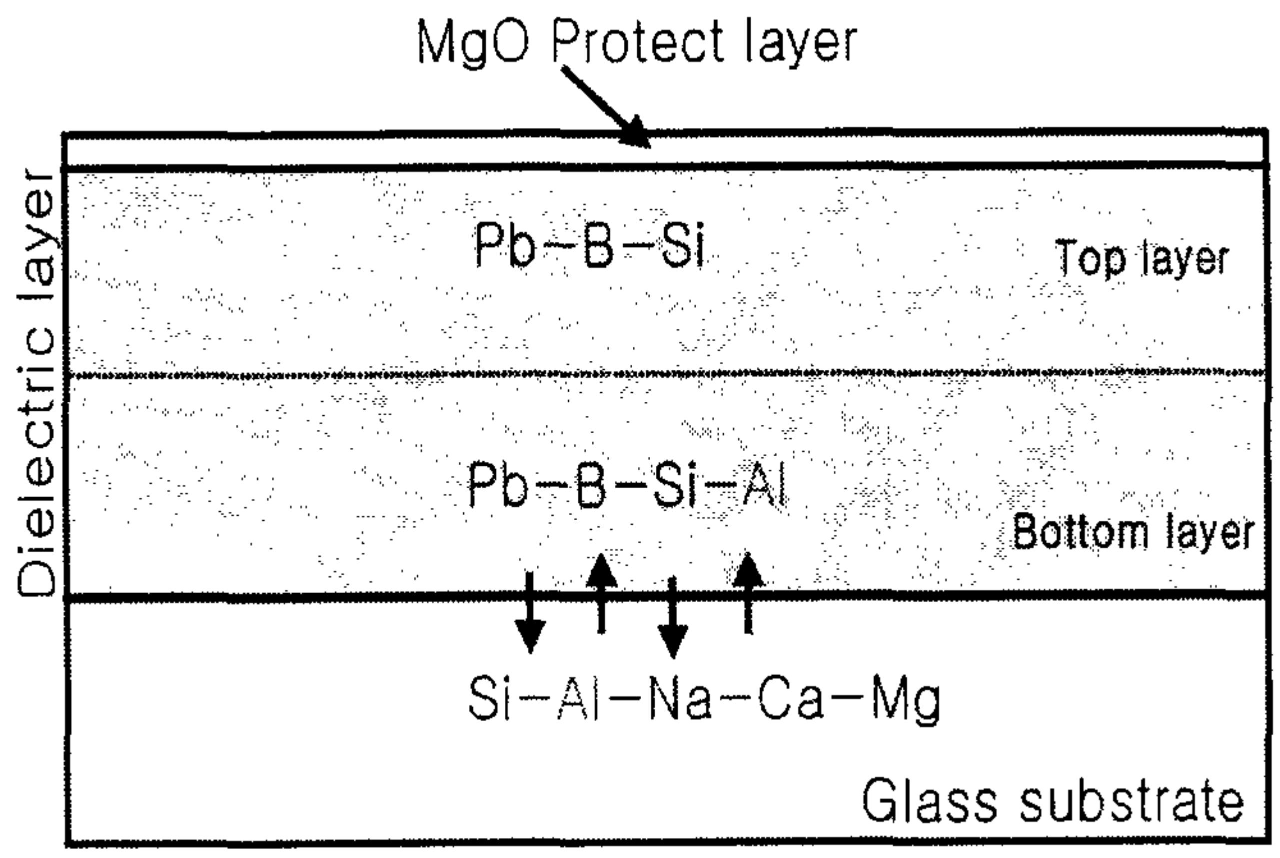

Fig. 1. The schematic of cross section of dielectric layer and substrate in PDP

Figure 1 shows the cross section of dielectric and substrate in PDP. The behavior of Al_2O_3 is important in dielectric layer in terms of viscosity of glass and ion diffusion during firing stage. Al_2O_3 contained in dielectric layer and substrate can easily move to each layer. Therefore Al_2O_3 can affect some properties of dielectric, especially electrical and thermal property.

Table 1 Compositions of PbO-B_2O_3-SiO_2-Al_2O_3 glasses with variable ratios

	Composition (wt%)					
Sample	PbO	B_2O_3	SiO_2	Al_2O_3	$RO(R_2O_3,RO_2)/Al_2O_3$	
S1	60	10	10	20	3	(PbO/Al_2O_3)
S2	65	10	10	15	4.3	
S3	70	10	10	10	7	
S4	75	10	10	5	15	
S5	60	10	10	20	0.5	(B_2O_3/Al_2O_3)
S6	60	15	10	15	1	
S7	60	20	10	10	2	
S8	60	25	10	5	5	
S9	60	10	5	25	0.2	(SiO_2/Al_2O_3)
S10	60	10	10	20	0.5	
S11	60	10	15	15	1	
S12	60	10	20	10	2	

Results and Discussion

The results of thermal expansion and dielectric constant on the $PbO-SiO_2-B_2O_3-Al_2O_3$ system are given in Figure 2-4. The relationship between the properties (dielectric constant and thermal expansion coefficient) and glass compositions were considered as a function of PbO/Al_2O_3, B_2O_3/Al_2O_3 and SiO_2/Al_2O_3 ratios.

Figure 2 shows the dielectric constants considerably increase with increasing PbO/Al_2O_3 ratio. PbO has highest dielectric constant (22.0 at 4.5×10^8 Hz) in comparison with B_2O_3 (3.0), SiO_2 (3.8) and Al_2O_3 (9.2), respectively. Although the thermal expansion coefficient of the glass is little sensitive to the change in the composition, it decreases gradually as the PbO/Al_2O_3 ratio increases.

Figure 3-4 show the thermal expansion coefficients and the dielectric constants are slightly decreased with increasing B_2O_3/Al_2O_3 and SiO_2/Al_2O_3 ratios. The original factors of each component to calculate thermal expansion coefficients from compositions are Al_2O_3 (16.67 at 20/100°C), PbO (13), SiO_2 (2.67) and B_2O_3 (0.33), respectively. As shown in the figures, the influence of Al_2O_3 on the thermal expansion coefficient is small.

The dependence of thermal expansion coefficient on B_2O_3/Al_2O_3 and SiO_2/Al_2O_3 ratio is consistent with the glass structure: the ionic radii of the ions Pb^{+2}, Al^{+3}, B^{+3} and Si^{+4} are 1.24, 0.51, 0.21 and 0.40Å suggesting that as B^{+3} has a greater strength of ionic field compared to Al^{+3}. Hence the glass structure containing B_2O_3 becomes stronger and is resulted in lower thermal expansion coefficient. The observed effects of SiO_2/Al_2O_3 on the dielectric constant of the glasses (S9-S12) agree with published data showing the similar trend [5]. The introduction of Al_2O_3, network modifier, raises the dielectric constant because of the formation of more easily polarizable non-bridging oxygen.

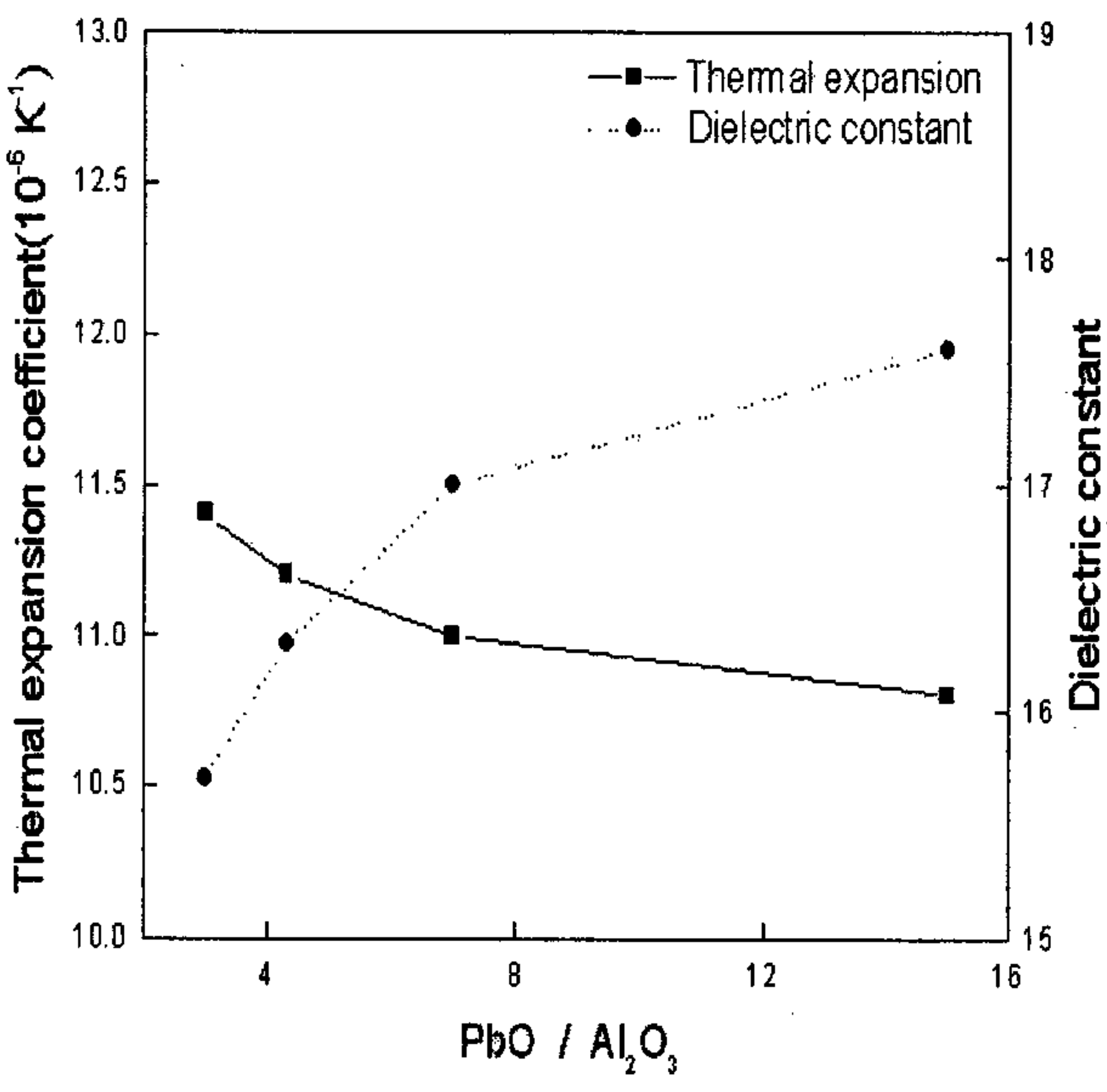

Fig. 2. Effects of PbO/Al_2O_3 ratio on the thermal expansion coefficient and dielectric constant of $PbO-SiO_2-B_2O_3-Al_2O_3$ (S1-S4)

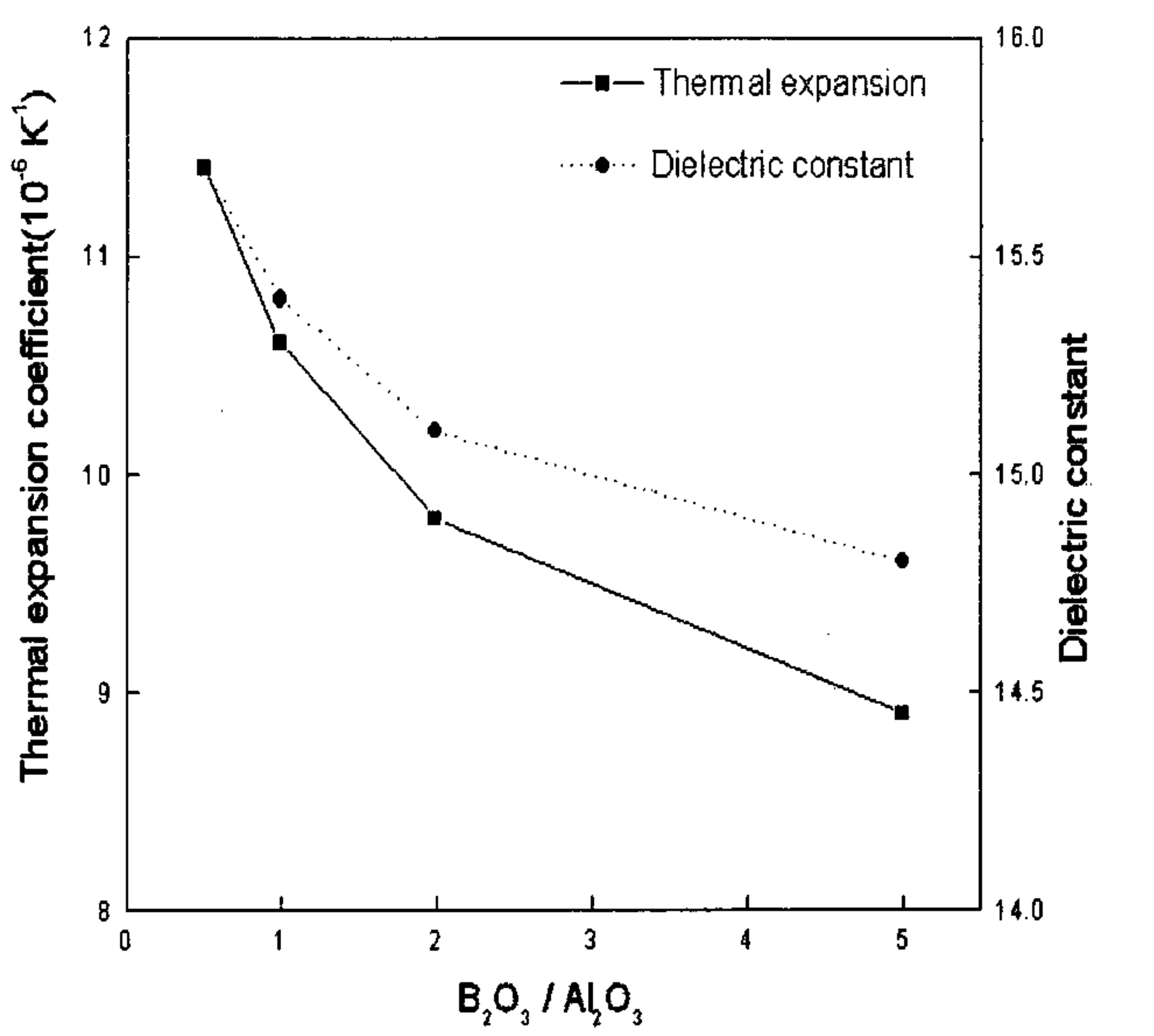

Fig. 3. Effects of B_2O_3/Al_2O_3 ratio on the thermal expansion coefficient and dielectric constant of $PbO-SiO_2-B_2O_3-Al_2O_3$ (S5-S8)

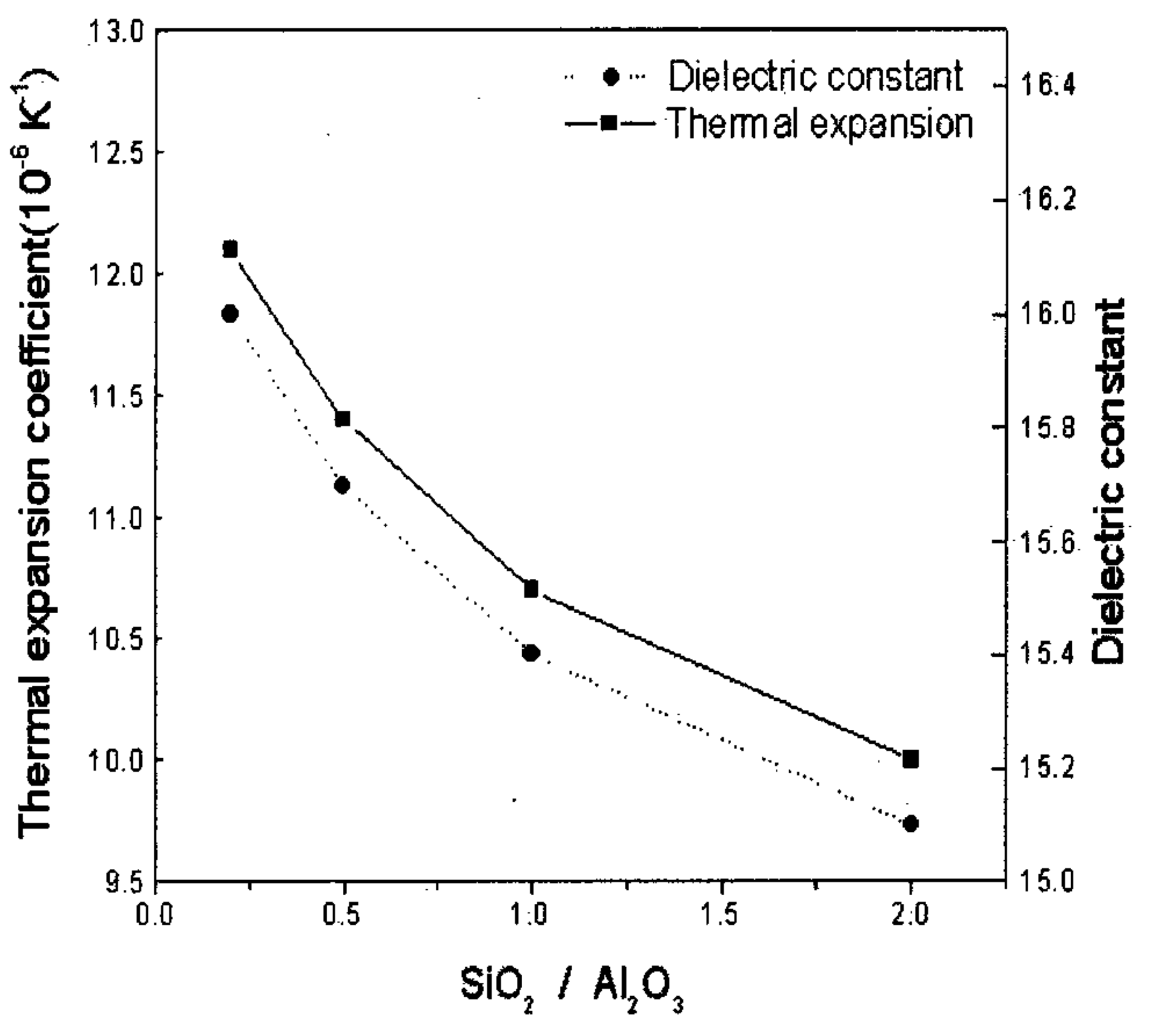

Fig. 4. Effects of SiO_2/Al_2O_3 ratio on the thermal expansion coefficient and dielectric constant of $PbO-SiO_2-B_2O_3-Al_2O_3$(S9-S12)

Conclusion

We researched the effects of Al_2O_3 in the $PbO-SiO_2-B_2O_3$ system by theoretical measurement. It was shown that thermal expansion and dielectric constant were greatly increased with increasing PbO/Al_2O_3 ratio, whereas B_2O_3/Al_2O_3 and SiO_2/Al_2O_3 ratios were little sensitive to the properties.

Reference

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