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Fabrication and characterization of B_2O_3–Al_2O_3–Na_2O glass system for transparent dielectric layer

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Abstract

Glasses within the B_2O_3–Al_2O_3–Na_2O system were examined as potential replacements for PbO-based glass frits with low firing temperature (500–600 °C) for transparent dielectric layer of plasma display panel (PDP). The glasses were evaluated for glass transition (T_g), softening temperature (T_f), thermal coefficient of expansion (α) and dielectric constant (ε). The dielectric constant of these glasses was also compared with theoretical data calculated by Appen and Bresker's equation. T_g of the glasses varied between 330 and 460 °C and T_f was in the region of 350–500 °C. The thermal coefficient of expansion ranged from 7×10^{-6} to 10×10^{-6} K^{-1}. The dielectric constant of these glasses was 6–8 within the region of the theoretical value. The T_g, T_f, α, ε of the selected glass, which was used to prepare transparent dielectric layer in 20 μm thickness, were found to be 449 °C, 476 °C, 8.284×10^{-6}/k, 6.84, respectively. All the results suggest that B_2O_3–Al_2O_3–Na_2O glasses would be suitable as a potential candidate for Pb-free dielectric layer in PDPs.

Keywords: C. Electrical properties; C. Optical properties; C. Thermal properties; Plasma display panel(PDP); Pb-free glass

1. Introduction

The plasma display panel (PDP), such as high-definition TVs and wall mounted TVs, has been much studied in the field of display devices due to large display area and wide viewing angle [1,2]. A transparent dielectric acts as a capacitor during electric discharge, restricts current, and provides a memory function in PDPs. It is formed on the front glass substrate by employing screen printing method to cover the display electrodes and bus electrodes. Generally, in order to develop a suitable transparent dielectric layer for PDP, such a film should satisfy several requirements: high transparency (above 80%), high break down voltage (above 4 KV), a low firing temperature between 550 °C and 580 °C, proper dielectric constant (< 15), and adequate thermal coefficient of expansion (8–9×10^{-6}/k) to match the soda-lime silica glass substrate [3,4].

In PDP, the PbO–B_2O_3–SiO_2–ZnO glasses have been widely used in commercial dielectric layers because of its high refractive index and low firing temperature below 580 °C [5–8]. However, with the enhancing awareness of environment protection, these materials containing PbO have been limited to prepare due to lead component deleterious to health and the environment. Therefore, low melting lead-free glasses have been extensively developed for PDPs. Many new lead-free systems, such as BaO–B_2O_3–ZnO [4,9], SnO–ZnO–P_2O_5[10], ZnO–B_2O_3–SiO_2[11], Bi_2O_3–B_2O_3–ZnO[12], BaO–Bi_2O_3–B_2O_3–ZnO [3,13–16] and Bi_2O_3–ZnO–BaO–Al_2O_3 [17,18] system, have been reported by a lot of research groups.

In this study, we characterized the thermal and electrical properties of B_2O_3–Al_2O_3–Na_2O glass system and prepared a new Pb-free transparent dielectric layer by exploiting this alternative frit composition for screen printing. At the meantime, theoretical data was used to compare with experimental properties.

2. Experimental method

The raw materials used in this research were chemically pure reagents without any further purification. All glasses were prepared

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with H_3BO_3 (99.5%, Sinopharm Chemical Reagent Co. Ltd., China), $Al(OH)_3$ (97%, Sinopharm Chemical Reagent Co. Ltd.), Na_2CO_3 (99.8%, Sinopharm Chemical Reagent Co. Ltd.). Organic vehicle were mixed with α-terpineol (95%, Aladdin, China), ethyl-cellulose (AR, Aladdin, China), tributyl citrate (98%, Aladdin, China), diethylene glycol monobutyl ether acetate (98%, Aladdin, China), Span85 (CP, Aladdin, China).

The glass composition is shown in Table 1. The well-mixed batches were melted in Platinum crucible at 1200–1400 °C for 1 h in air to achieve a homogeneous melt, and the melt was cast into a copper plate to proceed quenching. The resulted glasses were annealed at the temperature 10 °C above the glass transition temperature determined by differential thermal analysis (DTA) to relinquish the inner stress. The bulk glasses were cut and polished with 5 mm × 5 mm × 40 mm dimension to measure the thermal properties. The coefficient of thermal expansion (CTE) of each glass specimen was measured by a thermal mechanical analyzer (PCY-II, Xiangtan Xiangyi Instrument Limited Company, China) with a heating rate of 5 K/min up to softening temperature (T_f). The line CTE (α) was obtained as a mean value within the range between 50 and 300 °C. Glass transition temperature (T_g) and thermal stability was detected by differential thermal analyzer (Diamond TG/DTA, PerkinElmer, USA) under a heating rate of 5 K/min. The dielectric constant (ε) was measured by an impedance analyzer (4294-A, Agilent, USA), and was calculated from the capacitance values at 10^4–10^7 Hz. The surfaces of glass samples were coated with silver paste electrodes in a uniform area. The properties of the dielectric constant measured were compared with the theoretically calculated values by Appen and Bresker's equation (450 MHz) [19]. The transmittances of B_2O_3–Al_2O_3–Na_2O glasses were investigated by using a spectrophotometer (3101 PC UV–vis-NIR, Shimadzu, Japan).

The bulk glass was ground using a ball milling method for 12 h to produce glass powder. The mean particle size distribution was 3–5 μm. The glass powder was then mixed with organic vehicle by ball milling in order to make paste for screen printing. The glass paste was printed on a soda-lime silicate glass and dried at 150 °C for 15 min. The screen-printed glass substrate was fired at 400 °C for 15 min at a heating rate of 5 °C/min, and then at 580 °C for 30 min at a heating rate of 10 °C/min. The morphological characteristics including the surface and cross-section of the fired dielectric layers were observed by scanning electron microscopy (JSM-5900, JEOL, Japan). The transmittances of dielectric layers were investigated by using a spectrophotometer in the range of 200-1000 nm (3101 PC UV–vis-NIR, Shimadzu, Japan).

3. Results

Fig. 1 illustrates the approximate region for glass formation in the B_2O_3–Al_2O_3–Na_2O glass system. The open circles represent that the quenched and annealed melts of these composites can form transparent glasses. It is observed that the glasses in the region of B_2O_3–Al_2O_3 and Al_2O_3–Na_2O could not be made because of devitrification during melting. Only glasses in the region of B_2O_3–Na_2O form more favorably. Glass-forming region is located by the dotted line in the B_2O_3–Al_2O_3–Na_2O glass from previous work[20].

Thermal properties are measured for the samples without devitrification and the results are shown in Fig. 2 along with glass compositions. There is a parallel change for the glass transition temperature (T_g) and the softening point (T_f) when changing Na_2O at expense of B_2O_3 for constant Al_2O_3 contents. The T_g and T_f values increase with increasing Na_2O content as

Table 1
Compositions of B_2O_3–Al_2O_3–Na_2O glasses in this study.

Sample	Composition			**Notes**
	B_2O_3(mol%)	Na_2O(mol%)	Al_2O_3(mol%)	
BAN-1	90	5	5	Glassy
BAN-2	85	5	10	Glassy
BAN-3	80	5	15	Glassy
BAN-4	75	5	20	Devitrification
BAN-5	85	10	5	Glassy
BAN-6	80	10	10	Glassy
BAN-7	75	10	15	Glassy
BAN-8	70	10	20	Glassy
BAN-9	80	15	5	Glassy
BAN-10	75	15	10	Glassy
BAN-11	70	15	15	Glassy
BAN-12	65	15	20	Glassy
BAN-13	75	20	5	Glassy
BAN-14	70	20	10	Glassy
BAN-15	65	20	15	Glassy
BAN-16	60	20	20	Glassy
BAN-17	5	70	25	Devitrification
BAN-18	10	60	30	Devitrification
BAN-19	20	50	30	Devitrification
BAN-20	30	45	25	Devitrification
BAN-21	35	55	10	Devitrification

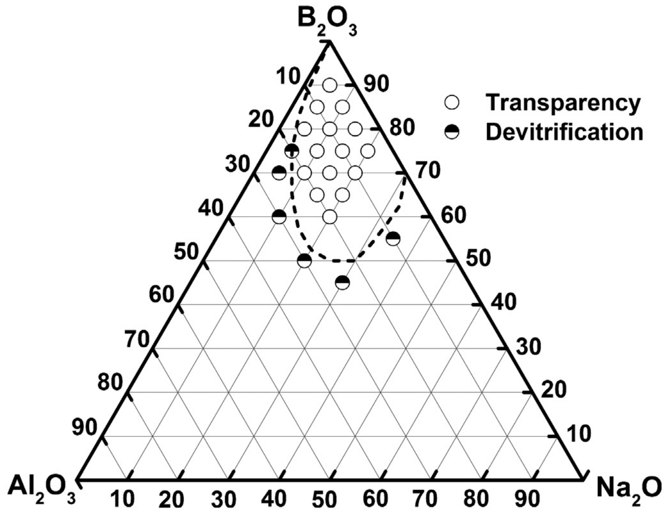

Fig. 1. Glass formation region in B_2O_3–Al_2O_3–Na_2O vitreous system (in mol%).

the amount of Al_2O_3 is below 15mol% (Fig. 2(a), Fig. 2(b)). As for 15mol% Al_2O_3, a slight increase in T_g and T_f is observed up to 10mol% Na_2O, followed by a fast decrease of higher Na_2O concentrations. The T_g and T_f values show a maximum around 10mol% Na_2O (Fig. 2(c)). This indicates that there would be certain structure changes around 10mol% Na_2O. While the content of Al_2O_3 is 20mol%, the T_g and T_f values decrease with the increasing of Na_2O(Fig. 3(d)).

The CTE results for the B_2O_3–Al_2O_3–Na_2O glass system are shown in Fig. 3(a), (b), (c), (d), respectively. When the amount of Al_2O_3 is below 10mol%, the CTE values of two series glasses increase from 6.931×10^{-6}/K to 9.884×10^{-6}/K (Fig. 2(a)) and 7.723×10^{-6}/K to 10.328×10^{-6}/K (Fig. 2 (b)) with increase of Na_2O content. The finding that T_g increases as the Na_2O content increases, whereas the CTE increases, is comparable to the results of Doweidar et al. [21] and Wakasugi et al. [22]. As for 15mol% Al_2O_3, the CTE value decreases from 8.653×10^{-6}/K to 7.633×10^{-6}/K with

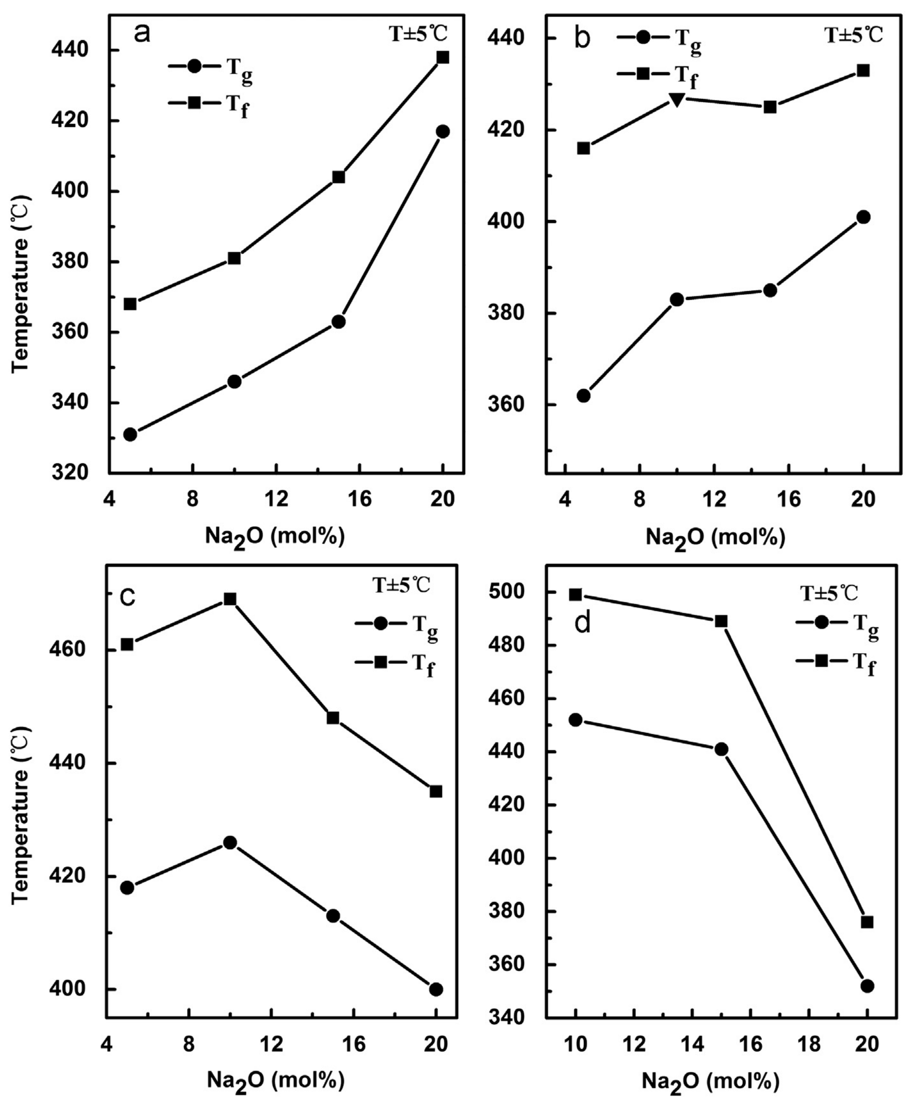

Fig. 2. Glass transition temperature (T_g), dilatometer softening point (T_f) as a function of Na_2O content: (a) 5mol% Al_2O_3, (b) 10mol% Al_2O_3, (c) 15mol% Al_2O_3 and (d) 20mol% Al_2O_3.

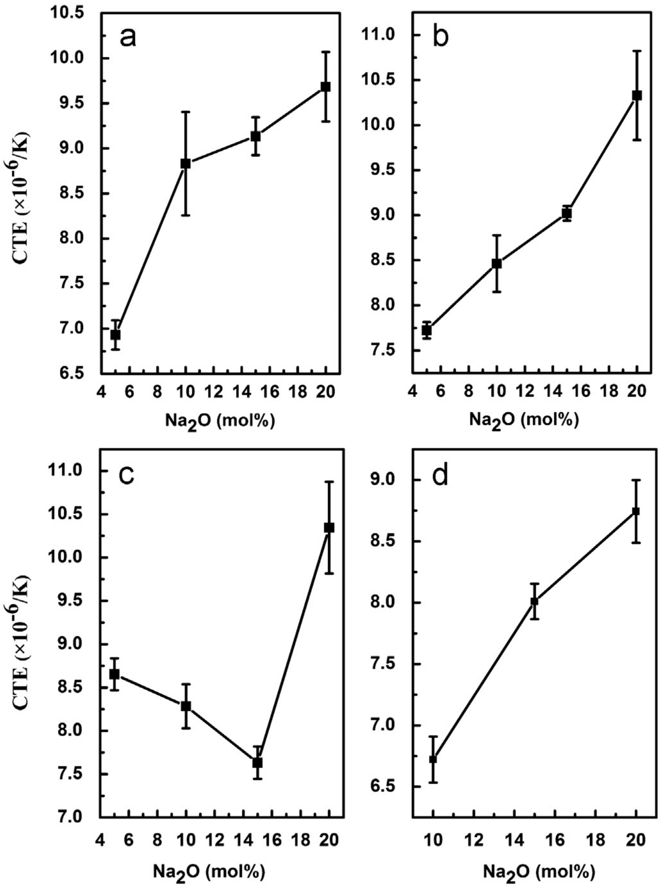

Fig. 3. Thermal coefficient of expansion (α) as a function of Na_2O content: (a) 5mol% Al_2O_3, (b) 10mol% Al_2O_3, (c) 15mol% Al_2O_3 and (d) 20mol% Al_2O_3.

the increment of the addition of Na_2O from 5mol% to 15mol %, and then increases to 10.345×10^{-6}/K when the addition of Na_2O is increased further to 20mol% (Fig. 2(c)). As the content of Al_2O_3 up to 20mol%, the variation tendency of CTE values is rising from 6.721×10^{-6}/K to 8.743×10^{-6}/K when the amount of Na_2O increases from 10mol% to 20mol% (Fig. 2(d)). In this condition, the compositional dependence in CTE shows opposite tendency to that in T_g and T_f.

Fig. 4 depicts the dielectric constant of B_2O_3–Al_2O_3–Na_2O glasses measured at 10 KHz, 1 MHz and 100 MHz. The dielectric constant remains relatively constant in the range of 10^4–10^7 Hz in this glass system, as the same result to BaO–ZnO–B_2O_3 glass system [4]. The dielectric constants are also calculated theoretically with the following equation described by Appen and Bresker [19]:

$$\varepsilon = 1/100\sum\varepsilon_i p_i \tag{1}$$

where p_i is the portion of individual oxides in mol% and ε_i represents the characteristic factor for each oxide: Al_2O_3 is 9.2, Na_2O is 17.6 and B_2O_3 is 3–8. Because the factor of B_2O_3 is dependent on glass composition, the theoretical values are calculated for the range 3–8 for B_2O_3 as shown in Fig. 4.

Table 2 summarizes physical, thermal, optical and dielectric properties of the glass named BAN-7 such as density, CTE, softening point, visible transmittance, dielectric constant. The T_g, α, ε of this glass are 449 °C, 8.284×10^{-6}/k, 6.84, respectively. These results indicate that the BAN-7 glass used in this study is suitable for transparent dielectric layer of PDPs.

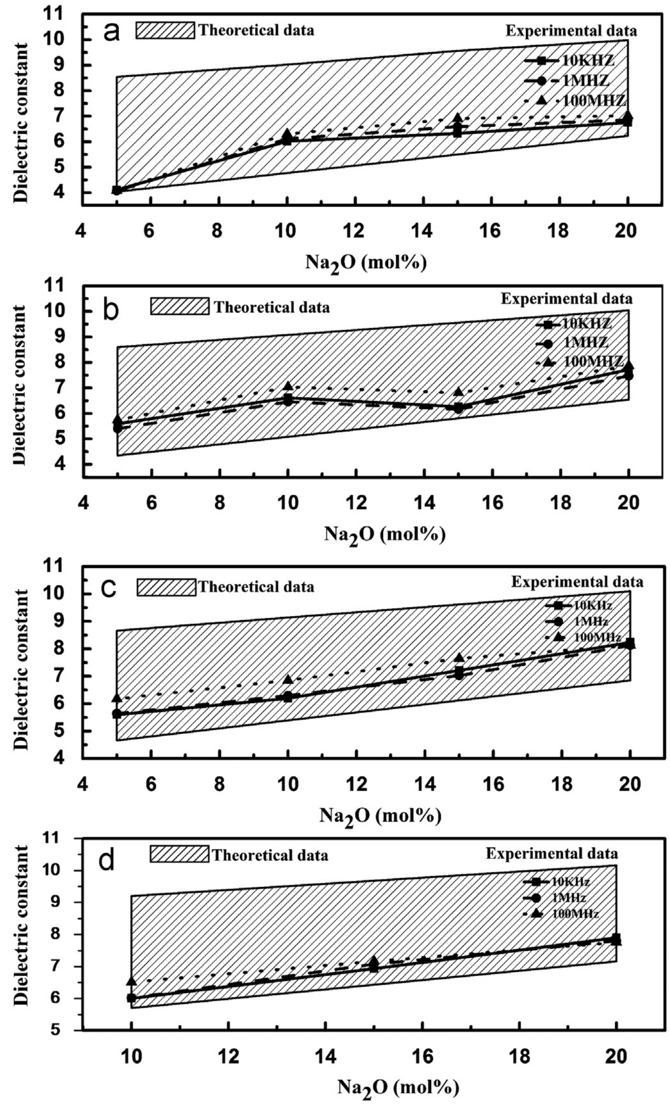

Fig. 4. Dielectric constant as a function of Na_2O content: (a) 5mol% Al_2O_3, (b) 10mol% Al_2O_3, (c) 15mol% Al_2O_3 and (d) 20mol% Al_2O_3 for B_2O_3–Al_2O_3–Na_2O glasses and comparison of experimental data (at 10 KHz, 1 MHz and 100 MHz) with theoretical data (at 450 MHz).

Fig. 5 shows the TG/DTA curves of the glass named BAN-7. No exothermic peak is observed for this glass, which indicates that BAN-7 glass shows amorphous stability without crystallization up to the melting temperature. In the TG curve, the weight loss of this glass powders is 8.43% at the temperatures below 600 °C.

Fig. 6 represents the surface and cross-sectional microstructure of the BAN-7 glass layer fired at 580 °C for 30 min. As shown in Fig. 5, there have been few pores existing in the surface and layer. The thickness of glass film obtained by heat treatment is about 20 μm.

Fig. 7 shows the transparency of the dielectric layers from the lead-free glass powder by the screen printing method. The glass film formed on a soda-lime glass panel exhibits a high

Table 2
Physical, thermal, optical, dielectric properties of the BAN-7 glass.

Density (g/cm^3)	CTE ($\times 10^{-6}$/K)	T_g (°C)	Transmittance (%)	ε
2.3684	8.284×10^{-6}/k	449	~90%	6.84

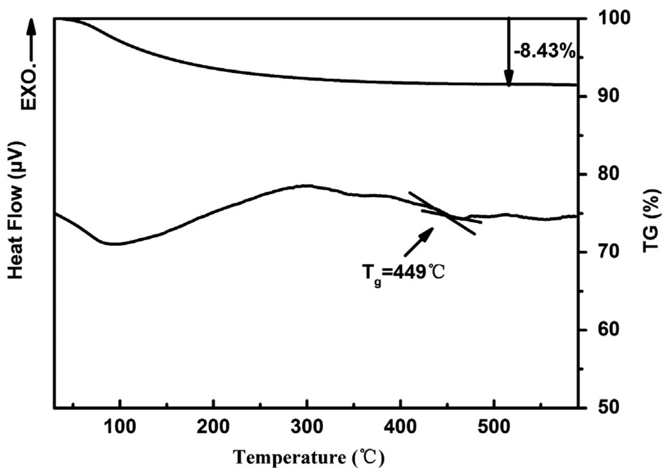

Fig. 5. TG/DTA curves of the glass named BAN-7 (heating rate: 5 K/min).

transmittance (~80%) in the visible region. As shown in Fig. 7, there has been almost the same appearance of high transparency between the glass layer and substrate.

4. Discussion

Conditions such as T_g, T_f, CTE, visible transmittance, and dielectric constant presented here are important factors in designing the glass composition for low firing temperature transparent dielectric layers in PDP [16]. In this study, the B_2O_3–Al_2O_3–Na_2O glass system can satisfy the various requirements for PDP without PbO additions.

The glass-forming area of B_2O_3–Al_2O_3–Na_2O glass system is smaller than Ref. [20] because of different cooling rate. The glasses in this work are annealed and quenched in air, while the glasses reported previously are quenched in air and water both. According to past research, Al_2O_3–Na_2O glasses and B_2O_3–Al_2O_3 glasses cannot be made. Only B_2O_3–Na_2O glass system form more favorably [20]. The results are in agreement with our work. So, fifteen glassy samples have been examined as potential replacements for PbO-based glass frits with low firing temperature.

In B_2O_3–Al_2O_3–Na_2O glass system, Na_2O associates with both Al_2O_3 and B_2O_3 to form AlO_4 and BO_4 units. For $Na_2O\leq 30$mol%, the fraction of four-coordinated boron ions (BO_4) is given as [21,23]:

$$N_{4(B)} = (1-0.031y)\frac{x}{100-x-y} \quad (2)$$

where x is the concentration of Na_2O in mol% and y is the concentration of Al_2O_3 in mol%. It can be concluded that the fraction of $N_{4(B)}$ increases with increasing Na_2O for constant Al_2O_3 contents. For $(Al_2O_3/R_2O)\leq 1$, Al_2O_3 has priority to associate itself with an equivalent quantity of Na_2O to form two AlO_4 units per Na_2O molecule. The T_g and T_f increase with the addition of Na_2O increasing from 5mol% to 20mol%, when the addition of Al_2O_3 is below 15mol%. It can be attributed to modifications of the network structure: the three-fold coordination of boron atoms in borate glass changes to four when alkali oxide is incorporate [24]. What's more, the concentration of Al_2O_3 in form of AlO_4 increases with addition of Al_2O_3 up to 10mol%. Since they are network forming units, the increase in the concentration of AlO_4 and BO_4 units can cause an increase in T_g and T_f. Once the content of Al_2O_3 is above about 15mol%, the main form is AlO_6 in B_2O_3–Al_2O_3–Na_2O glass system. The function of Al_2O_3 in form of AlO_6 is network modifiers and it induces a decrease of T_g and T_f. So, as 15mol% Al_2O_3 is added, the increasing in the concentration of BO_4 and the decreasing in the concentration of AlO_4 can induce a maximum of T_g and T_f at about 10mol% Na_2O. The T_g and T_f values decrease with 20mol% Al_2O_3 due to the decrease in the concentration of BO_4 and the accompanied increase in the concentration of AlO_6.

The addition of Al_2O_3 in B_2O_3–Na_2O glass system can improve the chemical stability and the ability of preventing devitrification from occurring. What is more, the addition of Al_2O_3 and Na_2O can also adjust the properties of this glass system, such as T_g, T_f, CTE. As can be seen in Fig. 2, T_g of the glasses varied between 330 and 460 °C and T_f was in the region of 350–500 °C. The T_g of glasses is all below firing temperature (550 °C–580 °C). As mentioned in Fig. 3, the thermal coefficient of expansion of these samples ranges from 7×10^{-6} to 10×10^{-6} K^{-1}. It is interesting to find that the CTE increases with T_g and T_s as well when adding Na_2O to the glass. The thermal expansion of the glasses is controlled by the amplitude of the thermal vibrations in the glass. It decreases as the rigidity of the glass network increases. An increase of the number of non-bridging bonds weakens the structure which in turn increases the coefficient of thermal expansion, whereas the change in coordination number of the network former cation may cause its increase or decrease depending on the effect on glass structure [25]. Here, the coordination number of cation has changed from 3 or 4 (BO_3 or BO_4 structure unit) to 4 or 6 (AlO_4 or AlO_6 structure unit). So it can induce an increase of the CTE. In this study, CTE increases with increase in the Na_2O content in the glasses which is shown in Fig. 3(a) and (b). The glass transition temperature is considered as an index of the chemical bonding energy between the atoms in the glass [26]. Addition of Na_2O into the glasses results in reshuffling the glass network. It forms the Al–O bond of AlO_4 tetrahedral unit and again rebuilds the structure by B–O–Al, hence the rigidity of the glasses increases. Therefore, the value of T_g increase when adding Na_2O in glasses. As a result, some

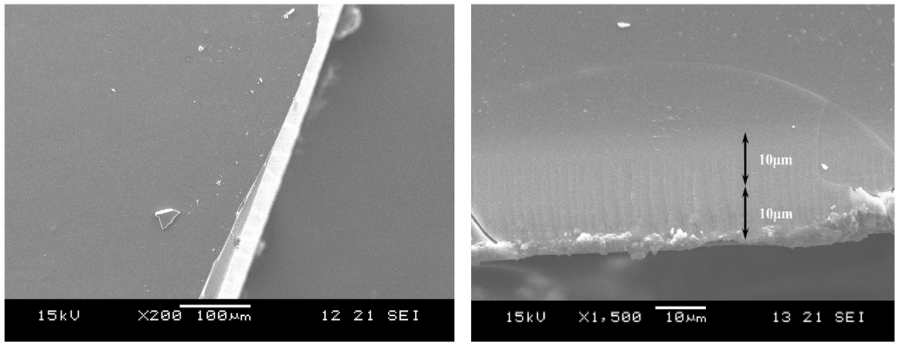

Fig. 6. Surface (left) and cross-sectional (right) SEM morphologies of the BAN-7 dielectric layer.

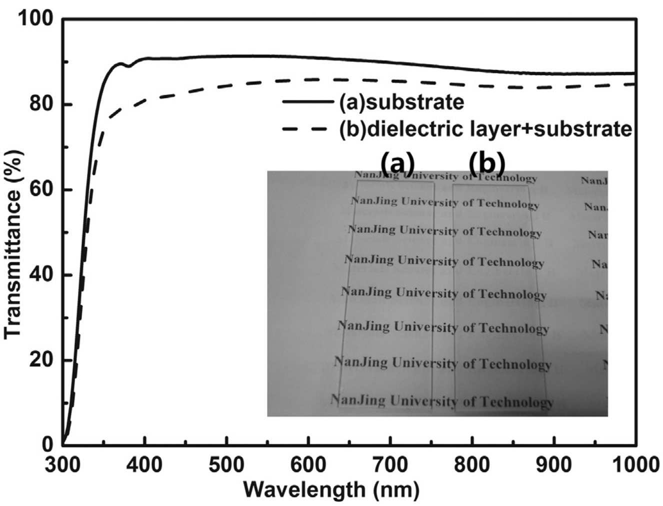

Fig. 7. Transparency of dielectric layer formed from the B_2O_3–Al_2O_3–Na_2O glass powders.

samples such as BAN-5, BAN-6, BAN-7 and BAN-10 can satisfy the requirement of CTE as transparent dielectric layer for PDPs because of adequate thermal coefficient of expansion ($8–9\times10^{-6}$/k) to match the soda-lime silica glass substrate.

The ε values of four series glasses increase as the Na_2O content increases. By comparsion, the ε values with high Al_2O_3 content are slightly higher than that of the glasses with low Al_2O_3 content at the same frequency. The reason for this variation may be that the deformability of sodium ions and aluminum ions is much greater than that of boron ions. The high polarizability of sodium ions and aluminum ions introduces high deformability ($Na^+=17.9$, $Al^{3+}=5.2$, $B^{3+}=0.3$) [27]. The dielectric constants obtained from the experiment are all in the region of the theoretical values calculated by Appen and Bresker's equation. For B_2O_3–Al_2O_3–Na_2O glass system, the equation has validity in region of these glass compositions. As for the frequency effect, no significant difference is seen in this glass system, although slightly higher ε values are represented at relatively lower frequencies. According to the experimental results shown in Fig. 4, the dielectric constants of the glasses in this study are all below 15.

Taking all the conditions aforementioned into consideration, the B_2O_3–Al_2O_3–Na_2O glasses can be suggested as a potential replacement for PbO glass frits for the dielectric layer in PDPs. Forming a dielectric layer on glass substrate, the paste prepared with BAN-7 glass powers ($10Na_2O$–$75B_2O_3$–$15Al_2O_3$) and organic vehicle is coated by the screen printing method. The transmittance of glassy layer is expected to depend on several factor, such as oxygen deficiency, surface roughness, and impurity center [28]. The reason for high transmittance of dielectric layer is mainly attributed that the defects such as pore and oxygen deficiency is relatively free. The transparent dielectric film, 20 μm in thickness, is obtained by proper heat treatment without few pores in surface and cross-section of layers.

We suggested B_2O_3–Al_2O_3–Na_2O glasses as a potential replacement for PbO glass frits, which cause environment problems, for the dielectric layer in PDPs. Although there are several requirements for the dielectric layer, we examined the fundamental optical, thermal and electric properties as initial step before applying them commercially. The transparent dielectric layer was formed on glass substrate by the screen printing method. Moreover, some conditions, such as the test contents (dielectric strength test and break down voltage test) and technological conditions (fired temperature and layer thickness), should be investigated on the glasses and films further.

5. Conclusion

Low firing glasses based on B_2O_3–Al_2O_3–Na_2O system have been investigated in order to replace the PbO-based glasses for dielectric layer of PDP. These glasses have T_g (T_f) < 500 °C, thermal coefficient of expansion of $7–10\times10^{-6}\,K^{-1}$ and dielectric constant of 6–8. All properties derived from the studied glasses match well with theoretically calculated values. The preferred dielectric glass composition in the B_2O_3–Al_2O_3–Na_2O system is: $B_2O_3=75$mol%, $Al_2O_3=15$mol% and $Na_2O=10$mol %. The transparent dielectric film is obtained with the selected glass by screen printing method.

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