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Development of transparent dielectric paste for PDP

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플라즈마 디스플레이용 투명 유전체 페이스트의 개발

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Abstract Plasma display panel is a potential candidate for HDTV, due to the fact that the screen size can easily be increased by a thick film technology. In this study, transparent dielectric materials which satisfied the requirements of dielectrics for PDP was developed using lead alumina borosilicate glasses. The Paste which had thixotropic behavior suitable for screen printing was made of this glass composition. The paste became more thixotropic as the particle size decreased. After firing, the cross sectional area of the thick film was analyzed by SEM. The voids in the thick film were removed using bimodal particle system. The dielectric thick film showed good adhesion characteristics.

요 약 플라즈마 디스플레이는 후막기술을 이용하여 화면의 크기를 늘리는 것이 쉽기 때문에 고선명 TV의 가장 유력한 후보이다. 본 연구에서는 플라즈마 디스플레이용 유전체의 조건을 만족하는 lead borosilicate 유리를 이용한 투명 유전체 재료를 개발하였다. 또한 이 유리를 이용하여 페이스트를 제조하였다. 페이스트는 스크린 프린팅에 적합한 요변성을 나타내었고, 입자 크기가 작아질수록 더욱 강한 요변성을 나타내었다. 열처리 후 후막의 파단면을 전자현미경으로 관찰하였다. 후막의 기공은 서로 다른 크기의 평균입경을 갖는 powder를 사용함으로써 제거 될 수 있었다. 소성된 후막은 좋은 융착 특성을 나타내었다.

1. Introduction

In this study, transparent dielectrics materials which could be used in PDP were developed. The dielectrics used in this experiment was lead alumina borosilicate glass. The demands for the dielectric glass are as follows. Dielectric constant must be 10~14 and optical transmittance must be higher than 80 % in visible range. The coefficient of thermal expansion must match to that of the glass panel to prevent cracking and thermal stresses. The softening point must be 50~100℃ lower than the firing temperature (530~600℃) [1]. Paste, which consists of dispersed functional phase in organic vehicles, was made of one of the developed glasses. The cross-section of the fired thick film was observed by SEM. The transmittance of fired thick film was measured. The microstructure of fired thick film was controlled by the different particle size functional phase systems in the paste.

2. Experimental procedures

2.1. Glass fabrication

All compositions were prepared from chemically pure 99.9 % PbO, 99.9 % B_2O_3, 99.9 % SiO_2, 99.9 % Al_2O_3. Alumina was added to prevent immiscibliity as the PbO-B_2O_3-SiO_2 ternary system had a wide immiscible area [2]. The weighed batch was mixed with a dry ball mill. The mixed powders were melted in an alumina crucible for 1 hours at 1100℃ and were quenched in distilled water for paste fabrication. Molten glasses were also quenched in a brass mold for thermal, electrical and spectroscopic analyses. All samples were annealed at 480℃ for 30 minutes and cooled to room temperature.

The glass phase was confirmed by a powder x-ray diffraction. WDX composition analysis was performed. The transition point, the softening point and the linear coefficient of thermal expansion were measured using the dillatomter (Netzch dillatometer 402) at the heating rate was 10℃/min. UV/VISIBLE spectrophotometer (Hitachi U-3410 spectrophotometer) was used to measure the optical transmittance of the polished glasses. Dielectric constant was measured by RF impedence/material analyzer (Hewlett Packad 4291A) at 2 MHz.

2.2. Fabrication of dielectric paste

The PD70 glass was suitable for dielectrics of PDP among fabricated glasses. So the paste was made from PD70 glass. Quenched PD70 glass was crushed in an agate mortar. After screening with 60 mesh sieve, the powder was pulverized in an attrition mill. The different mean particle sizes of 5.14 μm ans 1.43 μm. powders were obtained with different milling time. Three pastes were made from these glass powders. The first paste consisted of 5.14 μm glass powder. The second consisted of 1.43 μm glass powder. The third consists of the mixed powder of previous two (50/50 wt%). The solid contents were same among the three pastes. Organic vehicles consisted of a solvent binder plastisizer. Buthyl carbitol acetate and buthyl carbitol mixed solvent was used. This mixed solvent which decreased the evaporation rate prevented the viscosity degradation and dissolved the binder well [3]. Binder was ethyl cellulose n-type. Plastisizer was dibuthyl phthalate considering the compatibility with binder. Other organic additives such as thixotropic agent, dispersing agent were used. The organic vehicles and glass powder were premixed in a stirrer. Wetting occurred in this premixing procedure. Paste was mixed in a 3 roll mill after premixing. Three roll mill consisted of feed roll, center roll, apron roll. Each roll had a different rotation speed. Paste was forced on to a high shear stress. Agglomerates were separated and air bubbles in paste were removed [3]. Paste viscosity was measured using a rheometer (Haake, RV20 Plate type).

Paste was screen-printed onto the soda lime glass substrate which was used in PDP applications. Mask frame was 200 mesh stainless steel. Printed substrate was dried at 130℃ for 20 minutes. Firing temperatures ranged from 560℃ to 600℃. The fractured surface was analyzed by SEM. The transmittance of fired thick film was measured by UV/ VISIBLE spectrophotometer (Hitachi U-3410).

3. Results and discussion

3.1. Glass properties

The dielectric glasses composition in this experiments was summarized in Table 1. WDX analysis results were also listed. The Al_2O_3 content was increased due to the corrosion of the alumina crucible. The powder X-ray diffraction pattern was ·
ıg. 1. Typical x-ray pattern of glass was observed.

The dillatometer analysis results were given in Table 2. Transition point (Tg) and softening point increased, while the coefficient of thermal expansion (CTE) decreased as sample's number increased (SiO_2 contents increased and B_2O_3 decreased) at the range from PD10 to PD40. The softening point decreased as the contents of PbO increased.

The transmittance of glasses were shown in Fig. 2. PD10, PD20 and PD70 glasses had higher than 80 %

Table 1
Transparent dielectric glasses system for PDP (wt%)

	PD10		PD20		PD30		PD40		PD50		PD60		PD70	
	batch	anal	bath	anal	bath	anal	bath	anal	bath	anal	bath	anal	bath	anal
SiO_2	7.99	7.51	10.7	10.06	13.43	12.57	16.18	15.31	7.5	7.09	6	5.27	6	5.51
B_2O_3	35.79	33.43	32.85	30.69	29.88	27.91	26.88	25.11	33.5	31.66	32	28.23	29	27.69
PbO	53.42	49.92	53.65	50.03	53.88	50.21	54.11	50.35	57	53.86	60	52.4	63	58.67
Al_2O_3	2.8	9.14	2.8	9.22	2.81	9.31	2.83	9.23	2	7.39	2	14.1	2	8.13

transmittance in visible range. PD30, PD40 samples higher than 75 %. The transmittance increase slowly as the wavenumber increases in visible and infrared range. Otherwise the transmittance in ultra violet range is poor. Plasma display panel uses UV light to excite the phosphor in operations [4]. So UV light must be shut out. PD70 glasses shows 0 % transmittance below 300 nm wavelength. This glass had the preferable optical properties.

Table 3 showd the dielectric constant of glasses. Dielectric constant increased with the amount of lead oxide. Dielectric constant must be 10 to 14 for applications in PDPs. PD70 glasses are suitable for dielectrics in PDPs considering the softening point, the CTE, dielectric constant and optical properties. So paste was made from PD70 glasses.

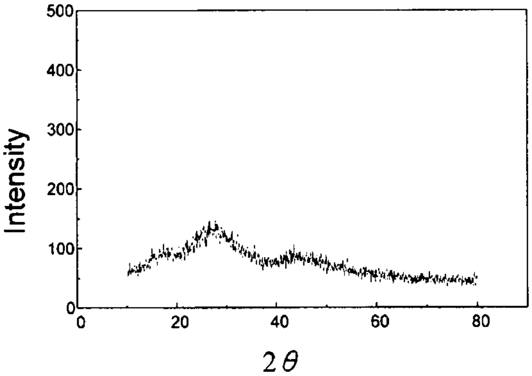

Fig. 1. X-ray diffraction pattern of glass.

Table 2
Thermal properties of glasses (CTE X 10^{-7}/°C)

Samples	T_g	Softening point	CTE (50~350°C)
PD10	476°C	510°C	81.720
PD20	469°C	512°C	75.455
PD30	484°C	518°C	46.773
PD40	490°C	530°C	38.687
PD50	471°C	503°C	82.225
PD60	480°C	510°C	73.801
PD70	472°C	497°C	60.222

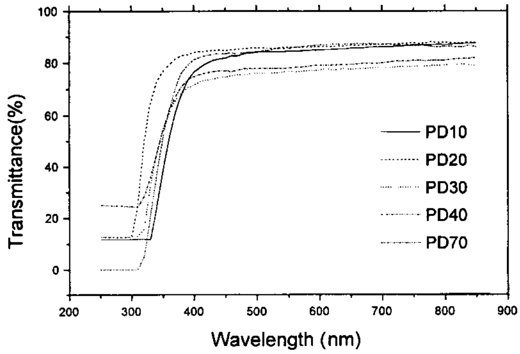

Fig. 2. The optical transmittance of glasses.

Table 3
Dielectric constant of glasses at 2 MHz

	PD10	PD20	PD30	PD40	PD50	PD60	PD70
Dielectric constant	10.08	9.96	9.92	9.82	11.39	11.21	12.35

3.2. Paste properties

Figure 3 showed the paste viscosity with the different particle size. All paste had thixotropic behavior and the paste showed higher viscosity and more thixotropic behaviors as particle size decreased. The surface area increases as the particle size decreases.Thus the paste viscosity increased as the particle size decreased at the same solid contents.

Figure 4 shows the optical transmittance of the

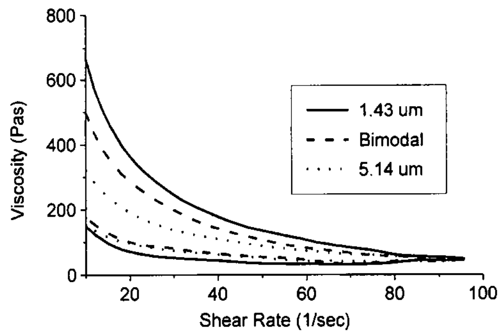

Fig. 3. The change of paste viscosity with different particle size.

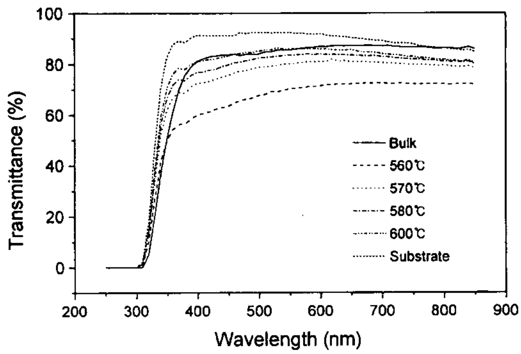

Fig. 4. The optical transmittance of bulk glass, substrate and fired thick films.

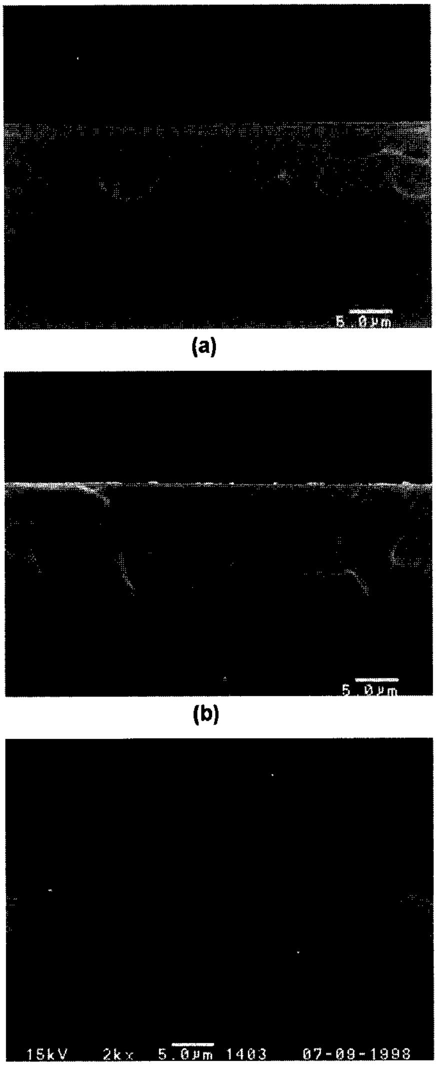

Fig. 5. The cross sections of fired thick films at 560℃ for 10 min.

fired thick film using the paste containing bimodal powders, bulk glass and soda lime silicate substrate. The optical transmittance of the fired thick film increased with the temperature because void was removed as the temperature increased. The optical transmittance of the thick film fired at 580℃ was higher than 80 %. The transmittance of thick film fired at 600℃ higher than 85 %. The more transmittance, the better brightness in PDPs. These thick films had preferable properties in UV range.

Figure 5 shows the cross-section of thick film fired at 560℃ for 10 minutes. The fired thick films using the paste containing 5.14 μm mean particle size powder and 1.43 μm mean particle size powder showed voids. However, the thick film using the paste containing mixture powders had submicron-size voids and showed good adhesion characteristics. This effects was due to the dense packing of powders during printing process. Therefore, using the bimodal system in paste was effective to remove the void at lower temperature. Figure 6 shows the cross section of fired thick film using bimodal paste at 580℃ for 10 minutes. Thick film fired at 580℃ showed no void and the smooth surface which was not obtained in the thick film fired at 560℃.

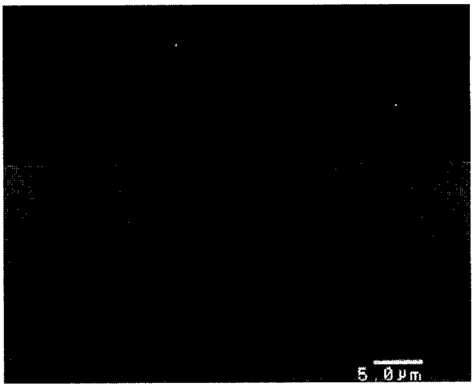

Fig. 6. The cross section of the fired thick film using bimodal paste at 580℃ for 10 min.

4. Conclusions

Transparent dielectric materials for plasma display panel were developed, using lead borosilicate glasses. PD70 glass was suitable for dielectrics in PDP. Softening point was 497℃ and dielectric constant was 12.35. The paste was fabricated using this glass. Using bimodal powder in the paste was effective method to remove the voids of thick film at lower temperature. The fired thick film at 580℃ shows no void, smooth surface, good adhesion to the substrate and higher than 80 % transmittance in visible range.

Acknowledgements

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