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Solvent Washing Dry Method for Aqueous Tape Casting

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For the fast dry of the aqueous tape, the process which water was replaced by organic solvent was proposed. So-called, it was the solvent washing dry. Three organic solvents (methanol, ethanol and acetone) were selected for the washing solvent. The weight loss of the washed tapes was measured to evaluate the dry rate of the tapes and dried tapes were examined the generation of the cracks with the variations of the organic solvent and the washing time. Methanol, ethanol and acetone were available organic solvents for this method. The tapes washed in methanol, ethanol and acetone were dried rapidly for twenty minutes. After thirty minutes, the weight losses were not found any more. The solvent of the lower surface tension can decreases the crack of dried tape. If solvent substitutes water completely, though it was fast dried, crack can be eliminated.

Key words: Organic solvent, Aqueous tape casting, PZT, Solvent washing dry

I. Introduction

Tape casting has been a normal method of production of thin ceramic sheets since its introduction by Howatt *et al.*[1] in 1947. This method is a low cost process for the manufacture of thin ceramic sheets with large areas of controlled thickness and high quality. Therefore, many kinds of ceramic components have been made using this method such as multi-layered ceramic capacitors, multi-layered ceramic packages and ceramic substrates.[2-6]

Tape casting has traditionally been performed using organic solvents as the liquid medium, but there is now a trend to move away from organic solvents and an expected transition towards water based system.[2] The main advantage of a water based system is that the health and environmental risks can be reduced. Lower cost and a minimized explosion risk are other advantages. The proposed major disadvantages of using water as a solvent are the slow drying rate and the flocculation due to strong agglomeration effects related to hydrogen bonding. [7]

The dispersion of the aqueous suspension is reviewed so much. The method to improve the dispersion mostly adds dispersant[8, 9] and controls pH. However, the drying rate of the aqueous tape casting has not been advanced until now. The increase of the drying rate in the aqueous tape casting may replace organic solvents to water in industry.

In this study, for the fast dry of the aqueous tape, the process which water was replaced by organic solvent was proposed. So-called, it was the solvent washing dry. Three organic solvents (methanol, ethanol and acetone) were selected for the washing solvent. For the evaluation of dry rate of the washed tapes, the weight loss of the washed tapes was measured. With the variation of the organic solvent and the washing time, dried tapes were also evaluated.

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II. Experiment

Commercial PZT powder was used in this experiment. PZT suspension was prepared by ball milling in deionized water for 24 h. The suspension was composed of 300 g of PZT powder (25 vol%), 30 ml of deionized water (30 vol%) and 1.5 ml (1 vol%) of dispersant (Duramax-3005, Rohm and Haas Company, USA). This process was used for all experiments for homogeneity and high solids content. Particle size distribution and specific surface area were measured with a rotational precipitation particle size analyzer (Shimadzu, SP-3, Japan) and a nitrogen adsorption surface area measurement based on BET method (Micromeritics, Flow Sorb II, USA), respectively. After dispersion, median particle size and specific surface area were 0.86 μm and 3.5 m^2/g, respectively.

PVA solution of 40 vol% (Poly vinyl alcohol, 10 wt%, Mw.=124000-186000, Aldrich) and PEG solution of 4 vol% (Poly ethylene glycol 400, Aldrich) were selected as a binder and a plasticizer. PVA solution and PEG solution were mixed and added to the dispersed suspensions with deionized water and mixed together. The slurries were mixed by slow ball milling for 3 h.

Within an hour, the three drops of the antifoamer (Sannop deformer 483) were added into the slurries, the slurries were de-aired by vacuum and stirring for fourty minutes.

Table 1. Physical Properties of Some Common Solvents for Tape Casting

Solvent	Surface Tension (erg/cm^2)	Dielectric constant (25°C)	Viscosity (mPa·s)	Boiling point (°C)	Evaporation rate (g/cm^2/s×10^6, 25°C)	Density, D^{20} (g/cm^3)
Acetone	25.1	20.7	0.306	56	5830	0.781
Ethyl alcohol	22.75(20°C)	24.3	1.074	78.4	1950	0.789
Methyl alcohol	22.5	32.63	0.544	64.6	3152	0.789
Water	72.75	78.54	0.89	100		1

The measured slurries were rapidly cast at three different thicknesses (the height of doctor blade) of 0.15 mm, 0.3 mm and 0.5 mm at a speed of 1.0 cm/s. The slurries were cast on silicon coated PET (Poly Ethylene Terephthalate) with a driven PET.

After casting, green tapes (casting height is 0.15 mm) put in the solvents for two minutes. The solvents were methanol (M-OH), ethanol (E-OH) and acetone. The green tapes that were washed in the solvents were measured weight loss by electric valance with drying time. In addition, in order to investigate dried tapes with the variation of the washing time, it was varied from 5 seconds to 120 seconds. Washed samples were dried in room temperature and the tape quality was evaluated.

III. Results and Discussion

The relative weights of the cast tapes are shown in Fig. 1. In this figure, the weight of tapes was decreased continuously and it needed about 4 hours in order to get the dried tape. The tapes washed in M-OH, E-OH and acetone were dried rapidly for 20 minute. After 30 minute, the weight didn't decrease any more. The weight losses of M-OH, E-OH and acetone were 22%, 33% and 24% respectively. These are due to the different evaporation rate of the washing solvents. For the washing solvent of the high evaporation rate, as soon as the cast tapes are washed, weight loss was so much before balancing those.

The drying times of the cast tapes washed by solvents were much shorter than that by water. That means solvent washing technique can be used for the fast dry in the aqueous tape casting systems. Table 1 shows the properties of solvent using in the tape casting. In this table, the evaporation rate of the solvents is compared with each other. The evaporation rate of acetone is the larger than those of M-OH and E-OH. However, the weight loss of E-OH is the larger than that of acetone.

The surface of the dried tapes is influenced by the surface tension of solvent. During drying the cast tape, solvent forms capillary between the particles of the tape. As the drying is progressed from surface to inner, a crack raises on the drying interface. In case of aqueous tape casting without substituting with solvent, there is no crack when the tape is dried at room temperature, but the tape is cracked when the drying rate is increased due to increasing temperature. In the Table 1, the surface tensions of solvents are shown.[10]

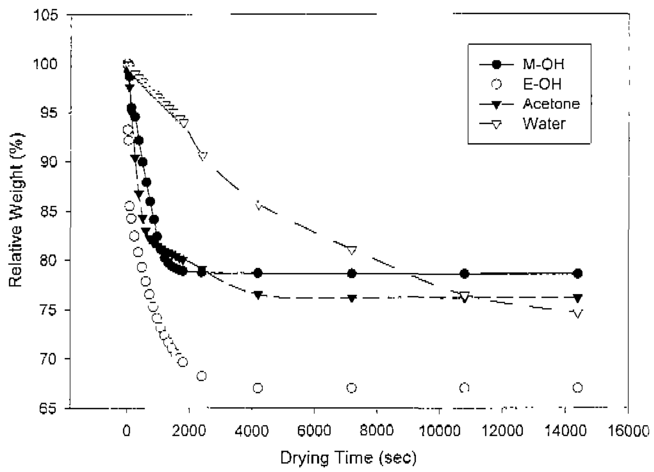

Fig. 1. The variations of the relative weight of the dried tapes with the variation of the drying time.

Table 2. The Defect of Green Tapes Dried by Methanol Washing with the Variations of the Washing Time and the Thickness of the Green Tapes

Tape Thickness	5 sec	10 sec	15 sec	30 sec	60 sec	120 sec
0.08 mm	O	O	O	O	O	O
0.18 mm	X	O	O	O	O	O
0.25 mm	X	X	X	X	O	O

O: Defect Free, X: Cracks

The surface tension of water is 72.75 erg/cm^2 and the higher than those of the others. In particular, it is the higher three times than those of M-OH, E-OH, and acetone used in this experiment. The decrease of the surface tension means the decrease of capillary force, stress, when the cast tape is dried. The solvent of the lower surface tension can decrease the crack of dried tape.

Table 2 shows the existence of crack on the PZT green tape washed by the M-OH with the variations of washing time and tape thickness. As the thickness of green tape increases, the washing time needed much more. When the thickness of green tape was 0.25 mm, it was needed a minute. However, in the case of 0.08 mm, it was needed just 5 seconds for the complete dried tape. The results of the E-OH and the acetone are shown in table 3 and Table 4. In the case of E-OH. the green tapes cast at the thickness of 0.25

Table 3. The Defect of Green Tapes Dried by Ethanol Washing with the Variations of the Washing Time and the Thickness of the Green Tapes

Tape Thickness	5 sec	10 sec	15 sec	30 sec	60 sec	120 sec
0.08 mm	O	O	O	O	O	O
0.18 mm	X	X	X	X	O	O
0.25 mm	X	X	X	X	X	X

O: Defect Free, X: Cracks

Table 4. The Defect of Green Tapes Dried by Acetone Washing with the Variations of the Washing Time and the Thickness of the Green Tapes

Tape Thickness	5 sec	10 sec	15 sec	30 sec	60 sec	120 sec
0.08 mm	O	O	O	O	O	O
0.18 mm	O	O	O	O	O	O
0.25 mm	X	X	X	X	O	O

O: Defect Free, X: Cracks

mm raised on the cracks. However, in the case of acetone, the cracks disappeared at the thickness of 0.18 mm. From the comparison of these table with the variation of the washing time and the solvent, it was known that acetone and M-OH were the similar effects for solvent washing drying, and decreased drying time of cast tape.

IV. Conclusion

In order to the fast drying of the aqueous cast tape, solvent washing method was used in this experiment. M-OH, E-OH and acetone were available for this method. The cast tapes washed in M-OH, E-OH and acetone were dried rapidly for twenty minutes. After thirty minutes, the weight losses were not any more. The solvent of the lower surface tension decreased the crack of dried tape. If solvent substitutes water completely, though it was fast dried, crack can be eliminated.

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