

## The Influence of Slip Characteristics and Casting Parameters on the Thickness of Thin Films Obtained by Tape Casting

S.E. Ivanchenko\*, I.O. Dulina, A.G. Nikulin, R.O. Kyrpal, S.O. Umerova, A.V. Ragulya

*Frantsevich Institute for Problems of Materials Science of NASU, 3, Krzhyzhanovsky St., Kyiv, 03142, Ukraine*

(Received 10 June 2013; published online 02 September 2013)

Tape casting slips with different composition have been prepared using BaTiO<sub>3</sub> nanopowder with a particle size in range of 20-25 nm. Ethanol, low polar organic solvent and menton were used as the solvents. Polyvinylbutyral and Butvar were used as binders. Dibutyl phthalate and polyethylene glycol where used as plasticizers. Rheological properties of BaTiO<sub>3</sub> suspensions were carried out on a rotary rheometer. Casted tapes have been characterized by optical microscopy and optical profilometry. Suspensions for tape casting characterized by pseudoplastic and pseudoplastic-dilatancy nature of the flow. The thickness of thin ceramic tapes depended on the height of doctor blade gap, the speed of the carrier and the viscosity of the suspension along with its rheological characteristics. Thickness of ceramic green tape in a greater extent determined by the casting parameters, composition of the suspension and its viscosity. The average tape thickness was 2.5 μm for low viscosity slips and near 5 μm for high viscosity slips with same blade gap in 50 μm. Roughness of tape depended on type of used solvent and its polarity and varies between approximately 500 nm for low polar solvent up to 1 μm for polar one.

**Keywords:** Tape Casting, Nanopowder, BaTiO<sub>3</sub>, Slip, Thin films, Multilayer ceramic capacitors, Viscosity, Rheological behavior, Pseudoplastic fluid, Thixotropy, Shear rate, Flow curve, Roughness of tape.

PACS numbers: 81.16.Nd, 77.55.+f

### 1. INTRODUCTION

Creating a modern, efficient materials of electric- and radio-technical purpose impossible without systematic scientific research in the fields of material science and new technologies. Especially, this applies to multilayer ceramic capacitors, for which the miniaturization becomes particularly important. Multilayer ceramics has many advantages in performance and reliability, cost and space saving. Tape casting is an important technology of ceramic processing, which are widely used in the production of thin sheets of flexible film. Tape casting is often the best choice because of its efficiency, relative simplicity and low cost in comparison with other available methods. The using of nanopowders in current technology allows to obtain thinner films with higher density. Therefore, the study of suspensions for tape casting based on nanopowders, and the resulting thin films is very actual for today.

### 2. MATERIALS AND METHODS

Tape casting suspensions have been prepared by using of BaTiO<sub>3</sub> nanopowder with mean particles size 20 – 25 nm, ethanol, low polar organic solvent and menton as the solvents, polyvinylbutyral and Butvar as binders, dibutyl phthalate and polyethylene glycol as plasticizers.

Suspensions of BaTiO<sub>3</sub> nanopowders have been characterized by rheological viscosity analysis and films were formed on casting machine TTC-1200 with a maximum carrier speed 55 cm/min. As the carrier was used politereftalat film coated with silicone adhesive coatings (Mylar). To identify the influence of casting parameters on the thickness of the film was used a

casting technique with changing the carrier speed and doctor blade gap during the casting process. Obtained tapes have been characterized by optical microscopy and optical profilometry.

### 3. RESULTS AND DISCUSSION

#### 3.1 Viscosity and rheological behavior of suspensions

From the results of flow curve measuring and the dependence of viscosity on shear rate has shown that the vast majority of slips for tape casting characterized by pseudoplastic dilatancy nature of the flow. Lack of dilatancy on flow curve was observed only with slips in which as an organic component was used ethanol and PVB and dibutyl phthalate as plasticizer.

Changing any of the components of the mixture resulted in the appearance of dilatancy and significantly decreased of the viscosity of the slip. From these results, the assumption can be made that for slips of BaTiO<sub>3</sub> nanopowders with low viscosity was typical to form an individual particle of barium titanate with binder, which has begun to interact with each other only at high shear rate.

Adding of dibutyl phthalate plasticizer caused some thickening of the slip on the one hand and usually led to appearance or strengthening of thixotropy on the other hand. Instead, adding polyethylene glycol caused almost no effect on the rheological properties of the slip.

Effect of solvent type was observed only by change of the viscosity of the system at low values of shear rate. Usage of less polar solvents in the suspension increased the viscosity of slip.

\* [sergonische@ukr.net](mailto:sergonische@ukr.net)

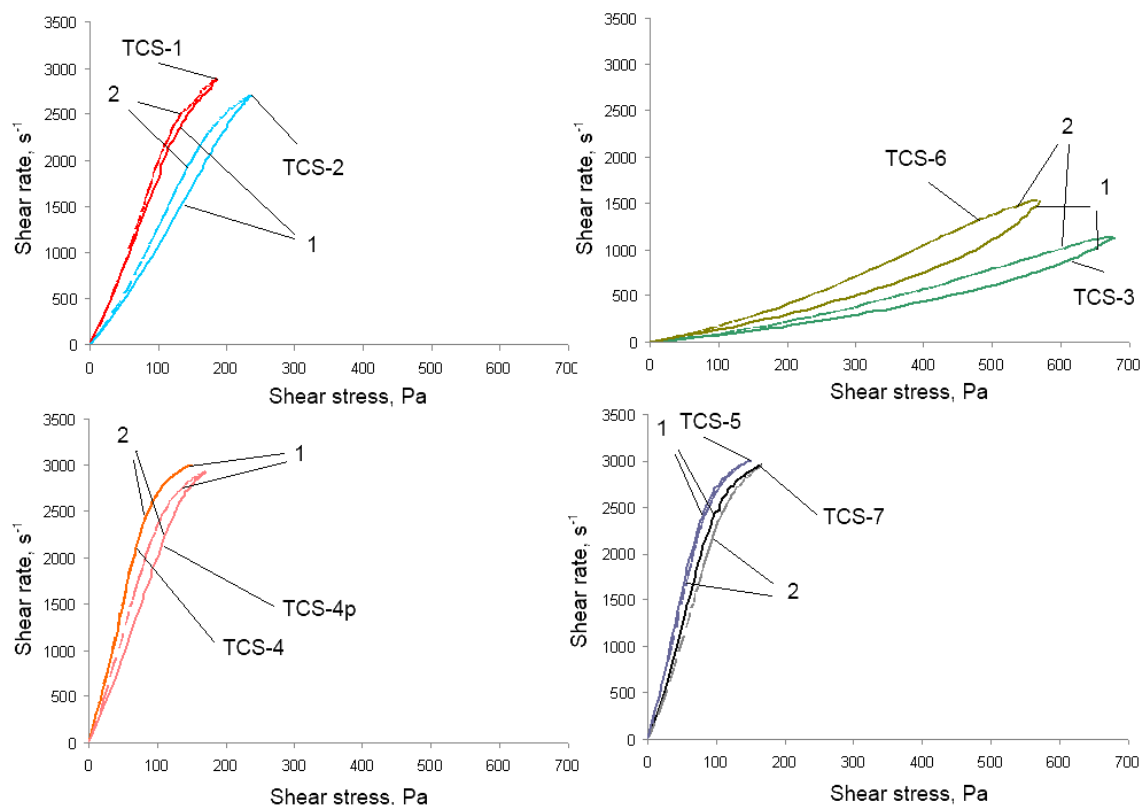
### 3.2 Influence of casting parameters on the thickness of the film

The influence of viscosity and rheological characteristics of the slurry on the thickness of the films showed that the wet film thickness obtained from slurries with

a viscosity of less than 800 mPa·s with using a small doctor blade gap mainly depended on the interaction of the rheological characteristics of suspensions and speed carrier.

**Table 1** – Composition of slurries for tape casting

Suspension name	Binder		Plasticizer		Solvent
	Material	Concentration, wt. %	Material	Amount, % wt. of PVB solution	Material
TCS-1	PVB (40000)	10	-	-	Menthone/ Ethanol (60/40)
TCS-2	PVB (40000)	10	DBF	1,5	Menthone/ Ethanol (60/40)
TCS-3	PVB (75000)	10	-	-	Ethanol
TCS-4	Butvar (40 000)	10	-	-	Ethanol
TCS-4p	Butvar (40 000)	10	DBF	5	Ethanol
TCS-5	PVB (750000)	5	PEG	5	Ethanol
TCS-6	PVB (75000)	10	DBF	20	Ethanol
TCS-7	PVB (75000)	5	DBF	20	Low polar organic solvent/Ethanol (Azeotropic mixture)



1 - forward curve 2 - backward curve

**Fig. 1** – Flow curves of suspensions for tape casting

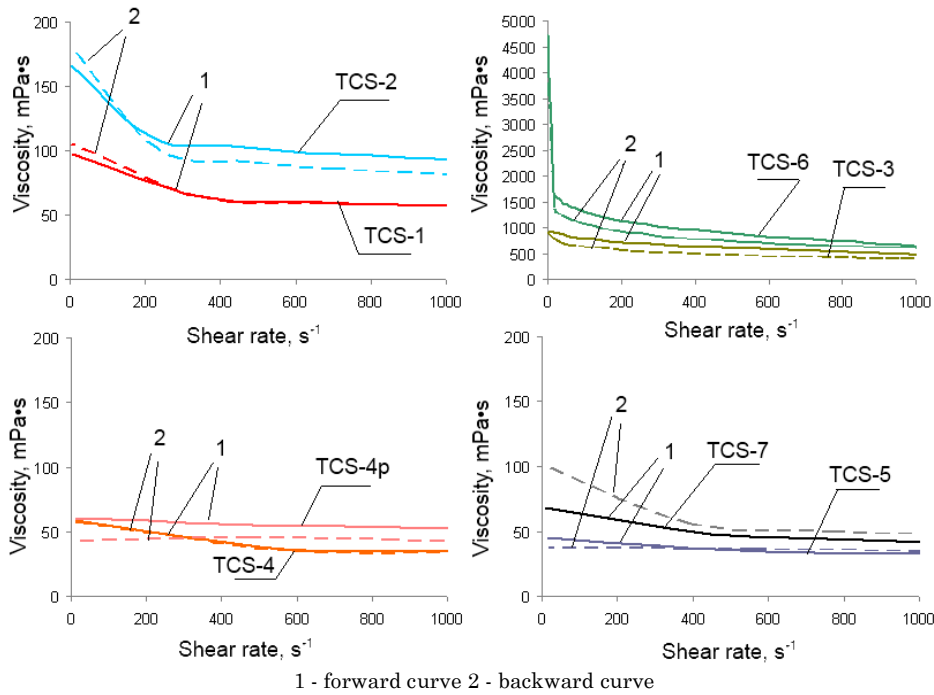


Fig. 2 – The dependence between viscosity and shear rate of suspensions for tape casting

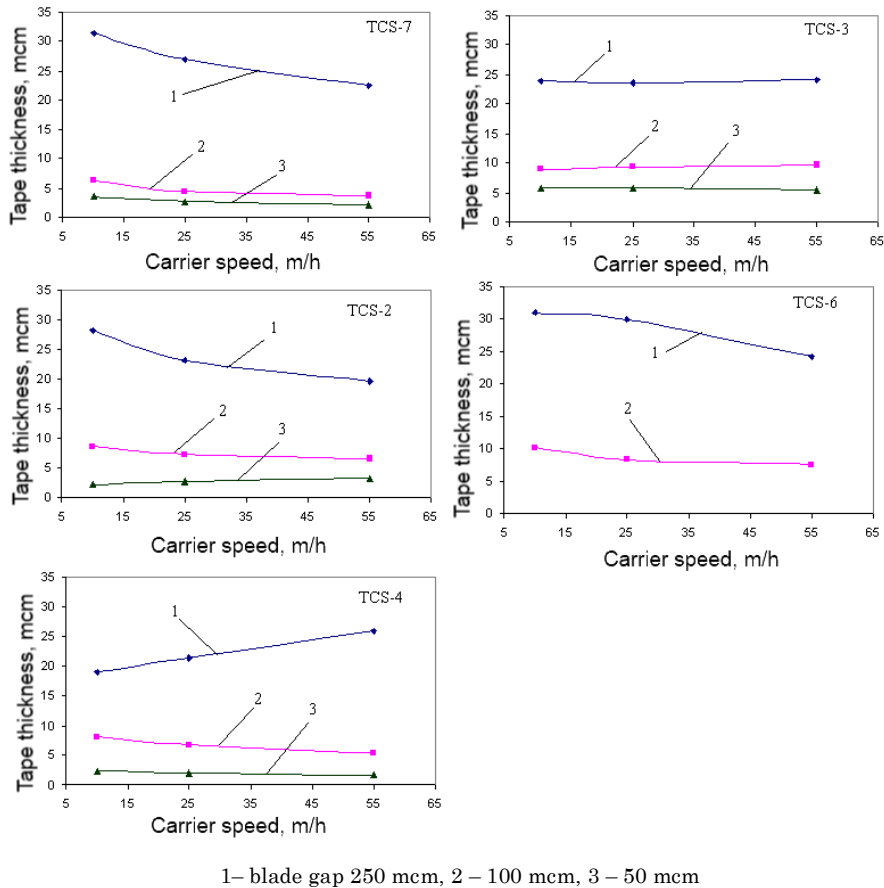


Fig. 3 – Influence of the carrier speed on film thickness obtained by tape casting with different doctor blade gap.

Suspensions with pseudoplastic and newtonian flow behavior had a linear dependence of the film thickness and height of the blade gap, but TCS-2 provided a thinner tapes by the same height of the blade gap be-

cause of it's low viscosity. TCS-7 had a lower viscosity and a dilatant flow behavior and showed good results at low height of the blade gap, but the film thickness increases at higher gaps, apparently due to a deca-

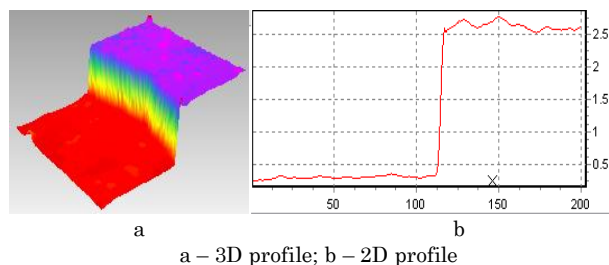
ing of mechanical action on it.

Changing of the carrier speed caused minor influence on the thickness of the tapes. TCS-7 responded to changes of carrier speed at all blade gap heights, because of its sensitivity to mechanical action. Wherein the thickness variation at small blade gap heights was observed in less responsive TCS-2. TCS-3 with pseudoplastic flow behavior hardly responded to changes in carrier speed, probably due to its high viscosity and thixotropic nature.

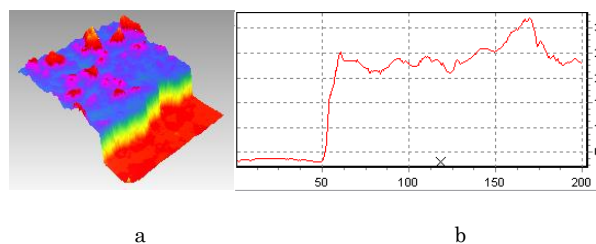
Increasing of thixotropic-pseudoplastic flow nature of the suspension increased tape thickness with increasing of carrier speed at small blade and the predominant pseudoplastic behavior of suspension led to thinning of the film.

### 3.3 Influence of solvent on tape roughness

Study of the profile of tape surface obtained from suspensions using low polar and polar solvents showed that the replacement of a polar solvent in the film on a low polar allowed to obtain films with less surface roughness. In our opinion, this is due to the fact that organic binders and plasticizers what has been used in the suspensions was low polar substances on the one hand and the polarity of the modified barium titanate powder surface caused the binder molecules join the plasticizer with polar groups and non-polar guide into solution on the other hand. The forming of such non-polar particles in suspension led to their separation from the polar solvent, resulting in "islands" of particles of the solid phase coated with a layer of organic binder what formed on the surface of tape.



**Fig. 4** – 3D and 2D surface profile of casted tape obtained from TCS-2 barium titanate suspension.



a – 3D profile; b – 2D profile

**Fig. 5** – 3D and 2D surface profile of casted tape obtained from TCS-5 barium titanate suspension.

It should be noted that based on the results of optical profilometry, this behavior of the particle of solid phase did not change the average thickness of the tape. The average tape thickness was 2.5  $\mu\text{m}$ . Roughness of tape varies between approximately 500 nm for low polar solvent up to 1  $\mu\text{m}$  for polar one.

## 4. CONCLUSIONS

1. Suspensions for tape casting based on barium titanate nanopowder characterized by pseudoplastic and pseudoplastic-dilatancy nature of the flow. The appearance of dilatancy observed only for low viscous suspensions at high shear rate.

2. On the thickness of thin ceramic tapes with equal composition has effect the height of doctor blade gap, the speed of the carrier and the viscosity of the suspension along with it's rheological characteristics.

3. In general, the thickness of ceramic green tape in a greater extent has been determined by the composition of the suspension and viscosity than by the rheological characteristics of suspensions for tape casting.

## ACKNOWLEDGEMENTS

Authors are grateful to the State Agency on Science, Innovations and Informatization of Ukraine for a financial support for the project NN352-2012.