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Tape Casting of Piezo Ceramic/Polymer Composites

Sven Schwarzer and Andreas Roosen*

University of Erlangen-Nuernberg, Department of Materials Science, Glass and Ceramics, Erlangen, Germany

Abstract

1-3 Piezoelectric composites are used as ultrasonic transducers for naval sonar devices, medical diagnostic systems, and non-destructive materials testing. Their excellent coupling behaviour to ultrasonic signals in water is due to the high fraction of polymer as the matrix and a very fine structured piezoelectric ceramic, which allows an effective transfer of the hydrostatic waves from the polymer to the ceramic. The aim is to obtain a composite of maximum hydrostatic performance characterised by d_h x g_h *(*d_h*: hydrostatic charge coefficient;* g_h*: hydrostatic voltage coefficient), which allows a detection of signals even with low intensity. Different processing routes are known to manufacture such 1-3 composites, but there is a need for a flexible, low cost process for medium quantities. Tape casting is a very adaptable method to produce such composites. Strips were cut into PZT green tapes line by line. Subsequently, the sheets were sintered and stacked, by using spacers between the sheets. The free spaces are filled with polymer, and finally the edges of the composite are cut off to remove the frame of the PZT-sheets. Different techniques were investigated to structure the green tapes without damaging them and to preserve the fine structure of the tapes during firing by avoiding warpage. © 1999 Elsevier Science Limited. All rights reserved*

Keywords: tape casting, composites, piezoelectric properties, PZT, piezoceramic/polymer composite.

1 Introduction

Piezoelectric materials find increasing demand in a wide field of applications within the last 40 years. As actuators, sensors, transducers etc. they are used in numberless technical systems. For the generation and detection of underwater ultrasonic waves, 1-3 composites have shown excellent properties and have convinced by their plain construction.[1] In this paper, a new method to produce piezoelectric 1-3 composites for transducer applications on the basis of tape casting is presented. Different processing routes are known to achieve fine scaled PZT ceramic rods, which are surrounded by a matrix of non-active polymer, such as epoxy.[2]

The technique of dice and fill[3] is based on a sintered monolithic ceramic block, produced by dry-pressing and sintering. This block is subsequently diced in crosswise by diamond saws. The ceramic rods which are obtained by this procedure are fixed in their position by remain standing on the non-diced block at the bottom. The filling of the cut block with a polymer, the removal of the non-diced area, the metallization of the front sides, and the polarization in an electric field leads to the 1-3 composite. The disadvantages of this process are long processing times per part and a reduced yield due to crack formation in the brittle ceramic.

By injection moulding[4] a ceramic green body with the typical rod-structure for 1-3 composites can be produced directly. After sintering, the parts run through the same procedure like the diced ceramic block. High costs for the injection moulding tools and the limited flexibility are the main disadvantages of this process. Tape cast composites were developed to avoid these disadvantages and to satisfy the demand for a flexible, low cost process.

2 Materials

A ceramic tape casting slurry (Table 1) based on lead-zirconium-titanate (PZT) was prepared.[5] In the first step the PZT powder, the organic solvents toluene and ethanol (66 vol%:34 vol%), and the dispersant were ball-milled for 24 h. In the second

*To whom correspondence should be addressed. Fax: +49-09131-8528311; e-mail: roosen@ww.uni-erlangen.de

Table 1. Slurry System

Components	*Typ*	*Vol%*
PZT	Ceramic powder	24·38
Tol/EtOH 66:34	Solvent	56·76
Hypermer CG6	Dispersant	1·92
PVB B98	Binder	10·13
BBP S 160	Plasticizer	6·70
Poly-Pale Resin	Compr. binder	0·11

step the binder system was added and dissolved in the solvent by ball milling for another 24 h. After degassing the slurry was cast on a silicon coated PET carrier film using the doctor blade process. The thickness of the green tapes could be varied between $\sim$10 μm to $\sim$2000 μm by changing the following parameters: the height of blade, the casting speed, and the viscosity of the slurry.

3 Processing

The 1-3 composite is based on two components, the ceramic rods and a polymer matrix. To form the rods ceramic green tapes were structured, sintered, and stacked using spacers between the sheets (Fig. 1). Square rods of 20 mm height, 0·7 mm diameter, and a distance to each other of 0·3 mm could be realised. Embedding the sheets with an epoxy resin, the composite is completed.

Figure 2 shows the complete processing route to produce 1-3 composites via tape casting. Strips were cut into the green tapes by three different methods: Copper-Vapour-Laser (CVL), YAG-laser, and by mechanical sawing. Especially the CV-laser is suitable to structure ceramic green tapes. A pulsed beam (max. 13 kHz) with low intensity (energy 1–25 mJ) allows to cut structures down to 50 μm width with submicron ranged heat-affected zones.[6] The structured sheets are sintered (T = 1180°C) and stacked. To produce complex 3-3 composites or thicker sheets, the green sheets are laminated prior to sintering. The free space is filled with polymer and finally the edges of the composite are cut off to remove the frame of the PZT sheets.

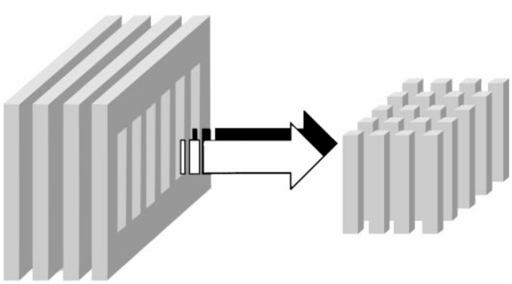

Fig. 1. Structural principle for the fabrication of 1-3 composites using structured ceramic sheets.

Now the composites are metallized with silver paste and polarized in an electric field of 2 kV/mm. After one day of ageing the piezoelectric properties were measured by impedance spectroscopy and berlincourtmeter.

4 Results

A comparison of the different methods to structure the green tapes showed that CVL is most effective to obtain high quality cut edges (Fig. 3). The YAG-laser energy input is too strong, the cuts are characterised by the appearance of heat-affected zones and typical melting noses. For mechanical structuring the applied binder system proved to be unsuitable, caused by its missing hardness and ductility. Only if sintered tapes were mechanically structured high quality cuts were obtained.

During sintering of the fine structured green tapes deformation occurred (Fig. 4), because of the different shrinkage behaviour in the areas (a), (b),

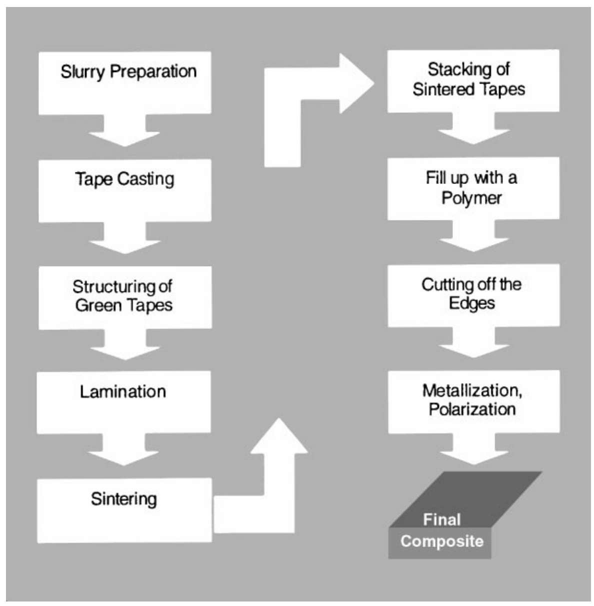

Fig. 2. Processing flow sheet of 1-3 composite fabrication.

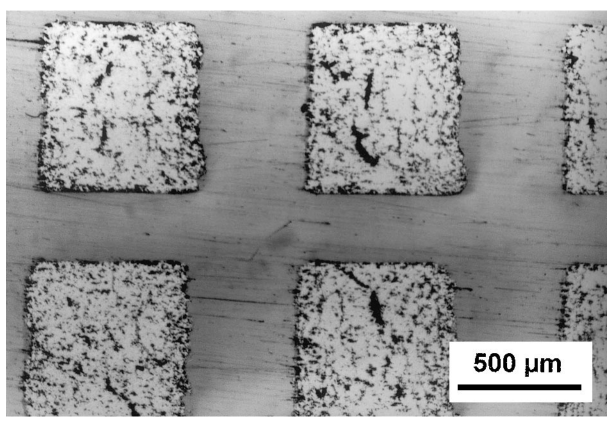

Fig. 3. Top view of a 1-3 composite structured by CV-laser cutting.

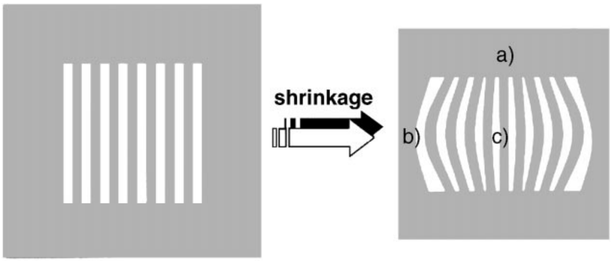

Fig. 4. Deformation of ceramic sheet during sintering.

and (c). This is due to the strips in the green tapes, which interrupt the ceramic structure vertical to the rods. To prevent the deformation during sintering, the sheets were sintered in a stack. The sheet at the bottom of the stack was a non-structured green sheet, which shows regular shrinkage. By adhesive power and friction, the above tapes also shrink regularly. Figure 5 shows 1-3 composites before metallization. The width of the cuts in a sheet (0,3 mm) and the distance between the tapes (1,0 mm) are not equal in this case. Table 2 summarises physical data of the pure PZT ceramic and the 1-3 composites.

5 Conclusion

A comparison of the processing routes for 1-3 composites produced by tape casting, dice and fill,

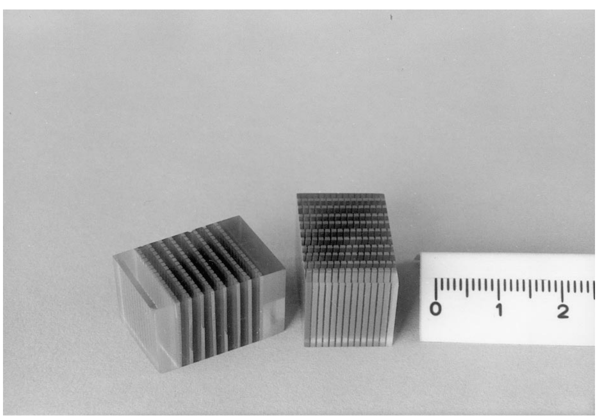

Fig. 5. 1-3 piezo ceramic/polymer composites.

Table 2. Properties of pure PZT ceramics and of 1-3 composites, produced by laser-cutting or sawing of ceramic tapes

Parameters	*PZT* SP53*	*1-3 Composites*		
		YAG	*CVL*	*Saw*
Density (gcm^{-3})	7·88	2·77	3·45	3·53
PZT (Vol%)	100	17.5	36	30
Permittivity	3800	660	1001	1146
tan d (%)	1·6	1·7	2·9	2·8
k_{eff} (%)	—	54	53	58
k_t (%)	51	58	56	62
d_{33} (pC/N)	650	395	409	466
g_{33} (mV m/N)	19·3	68	46	46

*CeramTec AG, Germany.

Table 3. Properties of 1-3 composites, produced by dice and fill

Parameters	*[7]*	*[8]*	*[3]*
Density (g/cm^{-3})	2·90	—	2·73
PZT (Vol%)	26	40	23
Permittivity	480	536	300
tan *d* (%)	1·7	—	2·9
k_t (%)	60	64	57
d_{33} (pC/N)	270	274	370
g_{33} (mV m/N)	65	58	140
$d_h g_h$ (10^{-15}m^2/N)	90	—	1820

–Unknown.

or injection moulding reveals the advantages of the new process. By achieving similar properties (compare Tables 2 and 3), the tape cast composite possesses advantages:

(a) cost-effective tape casting process;
(b) high flexibility in design by computer-controlled laser cutting of ceramic green tapes;
(c) high yield by contact-less laser-structuring;
(d) alternatively low cost cutting by mechanical structuring;
(e) possibility to manufacture complex structures and 3D parts by the lamination of green tapes;
(f) possibility to recycle waste of green tapes.

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