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07 MAY 2002

LAMINATED MULTILAYER THICK-FILM ACTUATOR**FIELD OF THE INVENTION**

5 The present invention relates to a process for producing multilayer actuators, and to such actuators.

BACKGROUND ART

 Piezoelectric ceramics have been accepted for various applications. Compared with ceramic pellet and bimorph structures, multilayer piezoelectric
10 actuators have several advantages, such as small drive voltage, quick response, large generative force, and high electromechanical coupling. The so-called tape casting method is the first choice for preparing multilayer piezoelectric ceramic actuators, as disclosed, for example, in US Patent Nos. 5,507,898 and 5,245,734.

 Generally, in this tape casting process, a slurry of piezoelectric powders, organic
15 solvent and binder is cast and molded into a green sheet on a tape using a doctor blade. Then a plurality of green sheets, each coated with an electrode, are laminated. After thermal compression bonding, the laminated body is sintered at high temperature.

 Screen printing technology, also known in the art as "thick film"
20 technology, is another method for preparing solid-state sensors and actuators. This technology has succeeded in manufacturing microelectronic circuits and simply structured electronic components and sensors. In thick-film printing the substrate, screen and paste come together at the screen printer. Generally a rubber squeegee blade traverses the top surface of the screen at a controlled
25 speed, the pressure causing thixotropic paste to flow through the pattern on the screen and hence onto the substrate. Snap-off and squeegee pressure affect the thickness of the wet print to a large extent. Too much pressure will cause spreading of the paste, whilst too little will result in an uneven print. After thick-



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07 MAY 2002

film printing, the substrate is allowed to stand in order to remove mesh marks, dried and then cofired with a specific profile. A paste of piezoelectric powders and an organic vehicle is forced through an aperture in a stencil screen and printed onto a substrate. Electrode and the piezoelectric layers are deposited alternately to form a sandwiched structure, and the printed films are sintered at a high temperature to form the so-called "thick-film" device. The device is commonly referred to as "thick-film" simply due to the processing method.

The main difference between normal tape casting and thick-film technology is that with the thick-film method the layers are formed from a wet paste and the amount of organics is at a much lower level. In contrast, with the tape casting method, the ceramic/electrode layers are laminated under dry conditions and the organic binder system in dried tape must have good handling properties, i.e. good elasticity, which unfortunately means a large proportion of organic binder is necessary and that therefore a large volume of pores are left after the organic binder is burnt off. In addition, the "thick-film" process can conveniently be used to prepare thinner piezoelectric layers than those prepared by the tape casting method, which means that a smaller voltage is required to drive the actuator.

Recent references about single piezoelectric "thick-films" include B. Morten et al, Hybrid Circuit 28 (1992), 25-28, Sensors and Actuators A31 (1992), 153-158, Microelectronics International 38 (1995), 5-11, IEEE Transactions on Ultrasonics, Ferroelectrics, and Frequency Control 43 (1996), 73-77; H.D. Chen et al, Journal of Applied Physics 77 (1995) 3349-3353; J.F. Fernandez et al., 30 (1995) 5359-5404. It is known that "thick-film" technology can also be used to fabricate multilayer piezoelectric actuators, as described by H.Moilanen et al in Sensors and Actuators A, Vol. 37-38 (1993) 106-111, Vol. 41-42 (1994) 421-425, Vol. 43 (1994) 357-365, and Microelectronics International 37 (1995) 28-30.

07 MAY 2002

However, significant technical problems still need to be solved to manufacture practical multilayer thick-film actuators. A critical issue is how to enhance the bonding strength between cofired internal electrode layers and piezoelectric layers and how to sinter the electrode and piezoelectric films to a reasonably high density without using too high a volume of sinter promotion additives. We have now found it possible to provide an efficient method to manufacture multilayer actuators, especially thick-film actuators, with compact structure and good performance. We have also found that this process may be applied to electrostrictive, as well as piezoelectric, ceramics.

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SUMMARY OF THE INVENTION

According to one aspect of the present invention there is provided a process for producing a multilayer actuator comprising the steps of:

- (a) forming alternate piezoelectric or electrostrictive ceramic layers and electrode layers on a substrate to form a laminate body comprising a plurality of piezoelectric or electrostrictive ceramic layers and a plurality of electrode layers, each electrode layer being interposed between two ceramic layers;
- (b) applying isostatic pressure to the laminate body; and
- (c) firing the laminate body.

Preferably the laminate body is placed inside a container to which the isostatic pressure is applied.

Preferably the container is in the form of a soft bag. Such a bag should be strong, i.e. have sound tenacity.

Preferably the layers are formed using screen printing, i.e. the process uses the so-called "thick-film" technique.

According to a particularly preferred embodiment there is provided a process comprising the steps of:

07 MAY 2002

- (a) forming alternate ceramic and electrode layers on a substrate to form a laminate body comprising a plurality of ceramic layers and a plurality of electrode layers, each electrode layer being interposed between two ceramic layers;
- (b) placing said laminate body into a flexible container, evacuating and
5 hermetically packaging the container;
- (c) immersing the container in a fluid medium, and applying compression to the fluid medium so that isostatic pressure is conveyed to the laminate body; and
- (d) firing the laminate body.

Thus, according to one preferred embodiment of the present invention
10 there is provided a process of producing multilayer thick-film actuators comprising the steps of:

- (a) screen-printing ceramic films and electrode films alternately onto a substrate to form a laminated body such that an electrode layer is sandwiched between ceramic layers;
- 15 (b) loading the substrate with said laminated body into a soft bag, and creating a vacuum inside the bag and hermetically packaging the bag;
- (c) immersing said hermetic package in fluid medium in a chamber, and applying compression to the fluid so that isostatic pressure is conveyed to the laminated body; and
- 20 (d) co-firing the compressed laminated body at high temperature.

Actuators of the multilayer type generally consist of a laminate block of alternate ceramic and electrode layers stacked over each other. The ceramic layer and electrode layer have to be deposited alternately so as to form the multilayer structure. Such a multilayer structure has a plurality of ceramic layers and a
25 plurality of internal electrode layers, which are laminated in an alternate manner, and at least a part of two adjacent ceramic layers has an internal electrode sandwiched therebetween. Each group of even and odd numbered electrodes, each of which is sandwiched between a couple of ceramic layers, is connected to

07 MAY 2002

a current collector. The current collector is generally in the form of an outer electrically conductive strip or layer applied on opposed side faces of the block perpendicular to the laminate. Preferably, an interdigital electrode structure, in which the electrodes with even number shift position in one direction with reference to the electrodes with odd number, is adopted. This structure is adopted in order that the odd electrodes do not short with the even collector on the opposite side of the block, or vice versa. Preferably, the electrodes may be staggered so that the length of the electrodes in each odd or even group is limited to be less than the width of the block so that the tip of each electrode is separated from the collector of the opposite group.

In a further preferred embodiment interdigital electrodes are achieved by using a stencil with two alignment marks to determine the position for printing the electrode layer. One alignment mark is used to align odd internal electrodes and the other for even electrodes. More preferably, a screen stencil used for screen printing the electrode layers comprises at least two sets of alignment marks, one for aligning the electrode layers with odd numbers with each other and the other for aligning electrodes with even numbers with each other. The odd and even electrodes are shifted in respect of each other by the difference between the two alignment marks. Efficiency of the process may be improved by printing more than one laminate body on a substrate. The laminate bodies and substrate may then be cut into separate actuators of desired shape. Thus, according to another preferred embodiment the laminate body is rectangular- or square-shaped and the laminate body is cut vertically, transversally to the stacked layers along two substantially parallel cutting edges such that one of the cutting edges only intersects the electrode layer with odd numbers and the other cutting edge only intersects the electrode layers with even numbers and after cutting, the electrode layers with odd numbers and the electrode layers with even numbers being separately exposed on opposite sides of the laminated actuator. It will be

07 MAY 2002

appreciated that a second substantially parallel pair of cutting edges, substantially perpendicular to the first, may be used to complete the rectangle or square shape.

Preferably, the inter-digital electrodes are connected by applying conductive paste on each of the sides with exposed electrode layers.

5 Thus preferably, the fired body together with the substrate is cut so as to expose the two terminals of the interdigital electrodes. Conductive paste is preferably used to connect all the odd internal electrodes at one terminal and all the even internal electrodes at another terminal, and then fired to form the external electrodes.

10 Preferably, a plurality of multilayer elements are deposited on one substrate in one multiple printing cycle so as to get a plurality of individual actuators after separation cutting.

 Preferably, the just deposited wet multilayer films are dried by heating before isostatic pressure is applied, preferably before placing into any flexible
15 container. Such drying preferably takes place at a temperature below about 200°C. At this temperature, generally any organic solvent has evaporated, but the other organic components, including organic binder, still remain in the laminate body.

 When a container is used, preferably, a vacuum is created inside the said
20 container with the multilayer film inside and then hermetically sealed. Isostatic pressure (hereinafter "IP") is applied preferably via a liquid medium. The adhesive strength between the electrode and the piezoelectric layers, and the density of the piezoelectric and electrode films, can be greatly improved by the isostatic compression.

25 In another embodiment of the IP process the compressive pressure is applied to the hermetic package and the sample inside via a gaseous medium or any other fluid-like medium.

07 MAY 2002

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In another embodiment, the isostatic pressure is applied after firing. Preferably firing takes place at a temperature above about 300°C. At this temperature the organic components including organic solvent and binder should be burnt off.

- 5 Preferably isostatic pressure is applied in the range of about 1,000 kg/cm² to about 10,000 kg/cm². Preferably the ceramic is a piezoelectric material, such as Pb(2r,Ti)O₃, but an electrostrictive material may be used. Preferably the electrode is Pd-Ag. The laminate body and substrate are then conventionally sintered to form the actuator.

10 **BRIEF DESCRIPTION OF THE DRAWINGS**

Various preferred features and embodiments of the present invention will now be described by way of non-limiting example with reference to the accompanying drawings, in which:

- 15 FIG. 1 shows a cross-sectional view of a multilayer piezoelectric thick-film element in accordance with one embodiment of the present invention;

FIG. 2 shows a view of one embodiment of a stencil which may be used to deposit the piezoelectric film (a) and the internal electrode film (b) to prepare the multilayer structure shown in FIG. 1;

- 20 FIG. 3 shows an embodiment of the printing and alignment sequence of the stencil marks for each piezoelectric and electrode layer which may be used to prepare the multilayer structure shown in FIG. 1;

FIG. 4 shows a schematic illustration of a cross-section of one embodiment of a multilayer body before cutting and it also shows the location of the cutting edges;

- 25 FIG. 5 shows a schematic illustration of one embodiment of the isostatic pressure process; and

FIG. 6 shows a further embodiment of the multilayer piezoelectric thick-film element with a printed PZT layer on the top of the multilayer body.

DESCRIPTION OF THE EMBODIMENTS

Preferred embodiments of this invention will be described below while referring to the drawings.

- As shown in FIG. 1, a multilayer thick-film actuator utilizing a piezoelectric effect according to the present invention has a laminated body 10. The laminated body 10 is formed by alternately screen-printed a plurality of internal electrode layers 11 and a plurality of ceramic layers 12 on an Al_2O_3 ceramic substrate 13; each of the layers is rectangular-shaped. Each pair of adjacent ceramic layers has one internal electrode layer sandwiched therebetween.
- 10 An interdigital electrode structure is preferably adopted, in which the electrodes with even number, 11b, shift slightly in one direction with reference to the electrodes with odd number, 11a. In this preferred embodiment, the ceramic layer 12 is formed of a piezoelectric ceramic consisting mainly of lead titanate zirconate, $\text{Pb}(\text{Zr},\text{Ti})\text{O}_3$ (hereinbelow referred to as PZT), with excess PbO added to promote the sinter process and to compensate for Pb loss due to volatilization at high firing temperatures. The internal electrode 11 is preferably formed from a palladium-silver (Pd-Ag) mixture. Each PZT layer preferably has a thickness of about 40 μm and each Pd-Ag electrode layer preferably has a thickness of about 5 μm .
- 20 The two edges of the rectangular-shaped internal electrodes 11, parallel with the cross-sectional plane in FIG. 1, are preferably covered completely with a PZT layer 12 (not shown), and the other two edges 14a and 14b, which are perpendicular to the cross-sectional plane, are exposed at opposite side surfaces of the laminated body 10. The exposed edges of the odd number internal electrodes 14a are preferably connected to each other by a film-shaped external electrode 15a, and the exposed edges of the even number internal electrodes 14b are connected by another external electrode 15b on the opposite side of the laminated body 10. The external electrodes 15a and 15b are preferably formed of silver
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(Ag). A pair of conductive leads 16a and 16b is preferably bonded to external electrodes 15a and 15b, respectively.

In this embodiment of the actuator, when a voltage is applied across the ends of leads 16a and 16b, it is applied via the external electrodes 15a and 15b to
5 internal electrodes 11a and 11b respectively, so that each ceramic layer 12 deforms. The deformations induced in the ceramic layers 12 are accumulated, so that the actuator induces a displacement in the laminating direction which is dependent on the applied voltage.

In general terms, when an electric field is applied the cations of the
10 ceramic are attracted in the direction of the field and the anions in the opposite direction, i.e. the ceramic layer expands or contracts in thickness.

In one preferred embodiment, to prepare the piezoelectric "thick-film" paste, fine PZT powder with a particle size preferably below 1 μm is mixed with
5 weight percent (%) lead oxide powder in a ball mill, and then a commercial
15 organic vehicle from Electro-Science Laboratory, Inc. (product #400 vehicle) is added in a weight ratio of ceramic powder: organic vehicle = 76:24, and the mixture is milled for 24 hours in a ball mill to improve its homogeneity. The use of the organic vehicle gives rise to a PZT-based piezoelectric paste has a suitable viscosity, rheology, and thixotropy for the following printing process.

20 For efficiency, preferably two screen printers are adopted to print the PZT film and Pd-Ag film, each through a stencil screen. FIG. 2a shows a preferred stencil pattern for printing the PZT layer. The rectangular area 23a is the open pattern in the screen. Viscous PZT paste is forced by a squeegee through screen apertures in the open patterns 23a to deposit the same pattern onto the substrate.
25 The blocked area 24a is coated with an emulsion film so that no paste can pass through. The cross mark composed of lines 21 and 22 is used for alignment. FIG. 2b shows a preferred stencil pattern for printing the Pd-Ag electrode film. The rectangular area 23b is the open pattern, through which the electrode paste

07 MAY 2002

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can be deposited with defined rectangular pattern. The area 24b is blocked with an emulsion film coat. The alignment mark is composed of two parallel lines 25 and 26, and another perpendicularly intersecting line 27. Preferably a screen with a thin emulsion coat and a fine mesh is used for printing the electrode film so as to achieve a thin final internal electrode.

A sequence of the multiple printing and alignment operation is illustrated in FIG. 3. Initially, by using one of the printers (hereinbelow referred to as printer A) with the screen stencil as shown in FIG. 2b installed, the first electrode layer E1 (also see FIG.4) is deposited onto the substrate 13 through rectangular elements 23b, together with the alignment mark 25, 26 and 27. This printed wet layer is dried at 170°C for 3 minutes in an oven. Then, the substrate with E1 is transferred to another screen printer (hereinbelow referred to as printer B) with the stencil as shown in FIG. 2a installed, and the first PZT layer P1 is printed. Each print is followed by drying in an oven. When printing P1, the screen marks 21 and 22 should be aligned with the marks 26 and 27, respectively, which were deposited on the substrate during the printing of E1. Each smaller rectangular electrode element 23b of E1 is completely covered by the corresponding larger rectangular PZT element 23a of P1. The substrate with E1 and P1 layers is transferred to the printer A again, and the second electrode layer E2 is printed with 25 and 27 aligned with 21 and 22, respectively, which were deposited on the substrate during the printing of P1. The positions of E1 and E2 shift slightly in respect of each other and by the same amount between 25 and 26 in the x direction as shown in FIG. 3. After E2 is dried, the substrate with the E1, P1 and E2 layers is transferred to printer B to deposit the second PZT layer P2 by printing with 21 and 22 aligned with 25 and 27, which were deposited on the substrate when printing E2. After P2 is dried, the substrate with E1, P1, E2, and P2 is transferred to the printer B, and the third electrode layer E3 is printed onto P2 with 26 and 27 aligned with 21 and 22, which were just deposited on the

substrate when printing P2. The alignment of E3 coincides with E1, and both E1 and E3 are slightly shifted with respect to E2 by the distance between 25 and 26 in the x direction. All the PZT layers, P1, P2, ..., are directly above each other.

The deposition of the PZT and Pd-Ag electrode layers may be repeated
5 alternately layer by layer until the desired structure is achieved. After completing the printing process, multilayer elements with an interdigital electrode structure, as illustrated in FIG. 4, are constructed.

The isostatic pressure (herein referred to as IP) process is schematically illustrated in FIG. 5. The substrate with a plurality of constructed elements is
10 loaded into a flexible container, preferably a soft plastic bag 31 with good tenacity. The soft plastic bag is evacuated using a vacuum pump and then the bag is hermetically packaged. The package is completely immersed in a liquid medium 32 in a chamber of an IP machine. The isostatic pressure 33 in the liquid 32 is built up to preferably 3500 kg/cm^2 at a preferred rate of $750 \text{ kg/cm}^2/\text{min}$
15 and the maximum pressure is preferably held for 10 minutes, and then pressure is released at a preferred rate of $600 \text{ kg/cm}^2/\text{min}$.

After removing from the plastic bag, the substrate with the plurality of multilayer elements is fired at preferably 400°C for 5 minutes in a belt furnace to burn off remnant organic binder, and sintered at preferably 1140°C for 10
20 minutes in a tube furnace. Then the substrate together with the fired multilayer elements is cut into individual actuators using a precision diamond saw. The cutting edge 17a only intersects the electrode edges with odd number (14a), whereas the cutting edge 17b only intersects the electrodes with even number (14b), as shown in FIG. 4 so that the terminals of the odd electrodes and the even
25 electrodes are exposed separately on opposite sides of the multilayer body 10. Conductive silver paste is used to connect all the odd electrodes and all the even electrodes on the two opposite sides, and then fired at preferably 700°C to form the external electrodes 15a and 15b, as shown in FIG. 1. Finally, a pair of

conductive metal leads, 16a and 16b is bonded onto 15a and 15b respectively to finish the preparation of the actuators.

As described hereinbefore, the IP process is important in achieving high quality multilayer actuators. Firstly, the bonding effect between the internal electrode layer 11 and the PZT layer 12 is greatly improved. Without the IP process, delamination is difficult to avoid, and this is greatly aggravated by mechanic tension resulted from the running diamond blade and resulting mechanic vibration during the cutting process. By applying the IP process to the laminated body, delamination is significantly decreased. Secondly, the density of the PZT layers is enormously improved. The contraction ratio (in thickness) of the compressed green multilayer body is 23% with respect to the dried green body before applying IP. After compression by the IP process and firing at 1140°C, the accumulated contraction ratio (in thickness) is 44% with respect to the thickness of the dried green multilayer before compressed. In contrast the contraction ratio after firing is only 28% for a sample to which the IP process is not applied. Therefore, with the IP process, the resulting actuator shows a much more compact structure and hence significantly improved performance.

Some of the main parameters for the above actuator are as follows: Room temperature dielectric constant of the PZT layer is over 600, and dielectric loss at 1 kHz is about 2%. Under a peak driving voltage of 150 V applied to a PZT layer with a thickness of 40 μm , the PZT layer shows a remnant polarization of 16 $\mu\text{C}/\text{cm}^2$ and a coercive field of about 23 kV/cm. After the samples are polarised with a voltage of 200 V in air, the PZT layer shows a d_{33} value over 600 pC/N.

A further embodiment of the above described process comprises applying the IP process to the multilayer body 10 after it has been fired at an intermediate temperature and the organics have been completely removed from the films.

Another embodiment of the actuator structure is illustrated as FIG. 6. The top of the actuator is covered with PZT layer 18 so as to prevent Pb volatilization at high temperature during the co-firing process. The PZT layer 18 can be formed by multiple printing.

- 5 The present invention is not restricted to PZT as the piezoelectric medium and Pd-Ag as the electrode. The same process can also be applied to the preparation of "thick-film" multilayer actuator composed of any other piezoelectric or electrostrictive ceramic material and any other metal electrode.

Claims

1. A process for producing a multilayer actuator comprising the steps of:

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(a) forming alternate ceramic layers of piezoelectric or electrostrictive material and electrode layers on a substrate both using screen-printing to form a laminate body comprising a plurality of ceramic layers and a plurality of electrode layers, each electrode layer being interposed between two ceramic layers;

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(b) applying isostatic pressure to the laminate body; and

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(c) firing the laminate body.

2. A process according to claim 1 comprising the steps of:

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(a) forming alternate ceramic and electrode layers on a substrate to form a laminate body comprising a plurality of ceramic layers and a plurality of electrode layers, each electrode layer being interposed between two ceramic layers;

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(b) placing said laminate body into a flexible container, evacuating and hermetically packaging the container;

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(c) immersing the container in a fluid medium, and applying compression to the fluid medium so that isostatic pressure is conveyed to the laminate body; and

(d) firing the laminate body.

3. A process according to claim 2 in which the flexible container is in the form of a soft bag.

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4. A process according to any preceding claim in which a plurality of laminate bodies are printed on one substrate, and the substrate and laminate bodies are then cut into separate actuators.

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5. A process according to claim 1 in which a screen stencil used for screen-printing the electrode layers comprises at least two alignment marks, one for aligning the electrode layers with the odd numbers with each other and the other for aligning electrodes with even numbers with each other, and such that the odd and even electrodes are shifted in respect of each other by the difference between the two alignment marks.

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6. A process according to any one of claims 4 or 5 in which the laminate body is rectangular- or square-shaped and the laminate body is cut vertically, transversally to the stacked layers along two parallel cutting edges such that one of the cutting edges only intersects the electrode layer with odd numbers and the other cutting edge only intersects the electrodes layers with even numbers, and after cutting, the electrode layers with odd number and the electrode layers with even layers are separately exposed on opposite sides of the laminate body.

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7. A process according to any preceding claims in which the laminate body is dried below 200°C before the isostatic pressure process is applied.

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8. A process according to any one of claims 1 to 6 in which the laminate body is fired above 300°C to remove the organic binder before applying the isostatic pressure process.

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9. A process according to any preceding claims in which the ceramic is a piezoelectric comprising $\text{Pd}(\text{Zr}, \text{Ti})\text{O}_3$ and the electrode comprises Pd-Ag, and the isostatic pressure is in the range of 1,000 kg/cm² to 10,000 kg/cm².

7 May 2002

16

10. A process according to any preceding claims in which a piezoelectric layer is printed on top of the laminate body.

5 11. A multilayer actuator prepared in accordance with the process of any one of claims 1 to 11.

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