J Electroceram (2006) 16:403–406
DOI 10.1007/s10832-006-9888-y

Impedance spectroscopy study and electrical behavior of PMN-PZT actuators joined by ceramic slurry

Jung-Hyuk Koh · Mun-Su Ha · Soon-Jong Jeong · Jae-Sung Song · Tae-geun Kim · Sang-Mo Koo · Jae-geun Ha

Abstract The solid solution of Pb(Mg,Nb)O_3-Pb(Zr,TiO) O_3 materials have high piezoelectric constant of 600 ∼ 650 pC/N and electromechanical coupling coefficient k_p of 0.65. Due to such high piezoelectric constant and electromechanical coupling coefficient, Pb(Mg,Nb)O_3-Pb(Zr,TiO)O_3 materials have been attracted attentions for the applications of multilayer ceramic actuators. Actuators can be produced by staking piezoelectric ceramic materials and inner electrodes, alternatively. However, it is difficult to fabricate huge ceramic actuators without any serious problems during the process conditions. Because ceramic are easily cracked during the sintering process, due to strikingly different shrinkage rate between the ceramic materials and metal electrodes. In this research, new jointing methods will be proposed for huge ceramic actuators, and then their electrical properties were investigated. Time dependent leakage current and impedance spectroscopy were employed to expect device performances.

Keywords PMN-PZT · Actuators · Spectroscopy · Green body joining

J.-H. Koh (✉)· S.-M. Koo · J.-G. Ha
Department of Electronic Materials Engineering, Kwangwoon University, Seoul, Korea
e-mail: jhkoh@daisy.kw.ac.kr

M.-S. Ha · S.-J. Jeong · J.-S. Song
Korea Electrotechnology Research Institute, Changwon, Korea

M.-S. Ha
Samsung Electro-Mechanics, Suwon, Korea

T.-G. Kim
Department of Electronics Engineering, Korea University, Seoul, Korea

1 Introduction

Significant progresses in functional ceramics and compound oxides for electronic device applications have been achieved in the past decade. Piezoelectric Pb(Mg,Nb)O_3-Pb(Zr,TiO)O_3(hereafter, PMN-PZT) [1–3] ceramics have perovskite structure with Curie temperature of 320°C. These PMN-PZT ceramics have high piezoelectric constant d_{33} of 600, and electromechanical coupling coefficient k_p of 0.65 as a ceramic form. Due to such high feasibilities of PMN-PZT ceramics, these materials have been attracted for the actuator applications. Actuators are devices, which can be turned on or off, adjusted or moved by stimulation. Generally, actuators use electrical signal or mechanical stress for their stimuli to response. Piezoelectric materials, which convert mechanical energy into electrical energy and vice versa, have been widely used for these actuators applications. Multilayer ceramic actuators have been commercially spread out for their large generation force and fast response. The overwhelming advantages of piezoelectric ceramic actuators can be summarized as fast response, large generation force, and low power consumption.

These ceramic actuators can be applied to the active isolation module and accurately controlled stage for probe station. It is extremely important to fabricate huge multilayer ceramic actuators without any cracks during the sintering process. However, as increasing the size of the actuators, cracks easily propagate along to the weak points generated by the stress [4]. To overcome this crack propagation and to fabricate huge ceramic actuators, many methods such as metal brazing [5–7], diffusion bonding [8], green state joining [9] have been proposed. These methods are not suitable for fabrication of multilayer ceramic actuators due to their different compositions. Green body joining might be an alternative method to build up huge ceramic actuators because it uses homogeneous composition with ceramic actuators. However, electrical and structural analysis for this method has not been yet presented.

In this article, we will present electrical characterization of green body joined PMN-PZT ceramic plate for actuator applications.

2 Experiments

In this experiment, tape casting method was employed to fabricate devices from the stoichiometric 0.2 $(PbMg_{1/3}Nb_{2/3})O_3$-0.8$(PbZr_{0.475}Ti_{0.525})O_3$ powder. PbO, ZrO_2, TiO_2, MgO, and Nb_2O_5 powders with high purity of 99.9% were used as starting materials. The powders were mixed and calcined at 850°C for 2 h and then slowly cooled down. 100 μm thickness green sheet were prepared through tape casting methods. 10 layers of green sheets were laminated for green body joining process. Finally green body with a dimension of 4 × 6 × 25 mm was cut.

Three different bonding types of ceramic were prepared to compare their electrical properties. Type (I) without green body joined ceramic (bulk ceramics), type (II) green body joined ceramics with different 2, 6, 12, 24 wt% binders, type (III) epoxy bonded ceramics were prepared to investigate their electrical properties. In all case, the size of ceramics plate is 4 × 6 × 25 mm. Ag electrode was applied to both top and bottom sides to analyze electrical properties. Electric field dependent polarizations were measured through Sawyer-tower method. Current (I)—voltage (V) characteristics, time dependent current behavior under constant electric field were tested through *Kiethley 6517A* electrometer. Finally complex impedance spectroscopy of PMN-PZT ceramic plates were investigated through *HP 4194 impedance analyzer*.

3 Results and discussion

Figure 1 shows electric field dependent polarization of different kinds of ceramics prepared by type (I) without interlayer (bulk ceramics), type (II) green body joined ceramics with different binder composition, and type (III) with epoxy bonded ceramics. To obtain field dependent polarization (Fig. 1), the spontaneous polarization versus electric field curves (P-E loop) curves of joined samples were measured by increasing electric field through a Sawyer-Tower circuit. In the figure, polarization of bulk and green body joined ceramics gradually increased near coercive field of 2.5 kV/mm. The increment of polarization increased drastically in further electric field except for the epoxy bonded sample. The bulk ceramics and green body joined ceramics show similar electrical behavior in polarization (P)—electric field (E) characteristics. However the epoxy bonded ceramics has different behavior. It seems that epoxy between the ceramic plates prevent polarization process under certain electric field of 3.5 kV/mm, and then polarization in epoxy bonded ceramics show similar polarization behavior with green body joined ceramics and bulk ceramics. This behavior probably comes from the weak dipole interaction of epoxy materials in the low electric field.

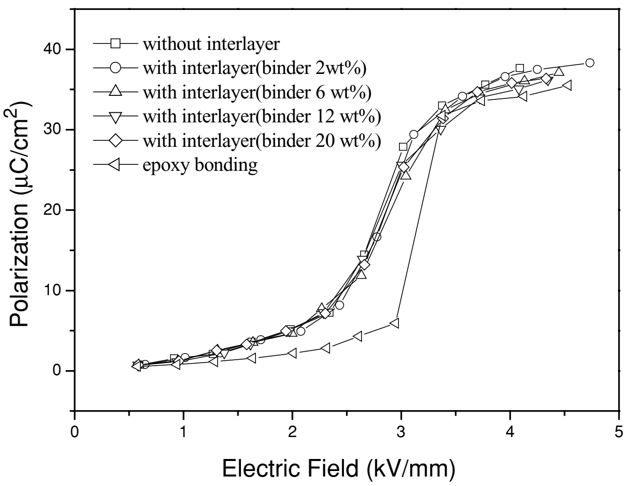

Fig. 1 Electric filed versus polarization of without interlayer (green body joined), with different composition of binders, and with epoxy bonded ceramics

Table 1 shows piezoelectric properties of joined (or bonded) samples with different joining (bonding) condition. As shown in the table, electromechanical coupling factor and piezoelectric coefficient of actuators joined by ceramic slurry have very similar to those of bulk actuator. Piezoelectric properties of epoxy bonded actuator became bad remarkably. When mechanical force and electric field applied to the bonded actuators, the epoxy layer in bonded part absorbs the applied mechanical force and electric field. Therefore, piezoelectric constant in epoxy bonded actuators have 40% decreased piezoelectric constant d_{33} compared with those of green body joined actuators and ceramic bulk actuators.

Table 1 Piezoelectric properties of different kinds of actuators

	Electric mechanical coupling factor, k_p	Piezoelectric consatant, d_{33} (pC/N)
Bulk ceramic actuator	0.31	419
Green body joined actuators (binder %)		
2%	0.31	414
6%	0.31	420
12%	0.31	414
20%	0.31	413
Epoxy bonded actuator (E)	0.24	246

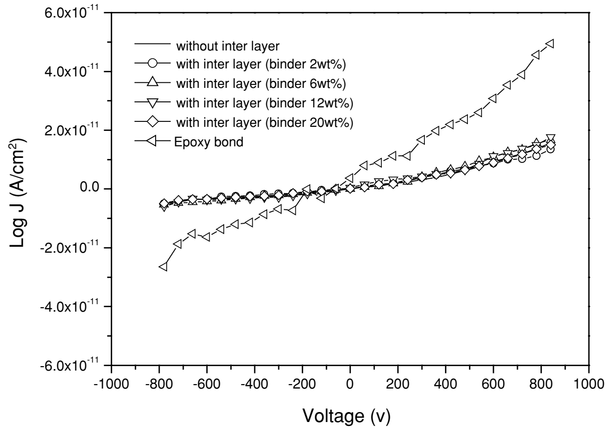

Fig. 2 Current (I)—voltage (V) characteristics of without interlayer (green body joined), with different composition of binders, and with epoxy bonded ceramics

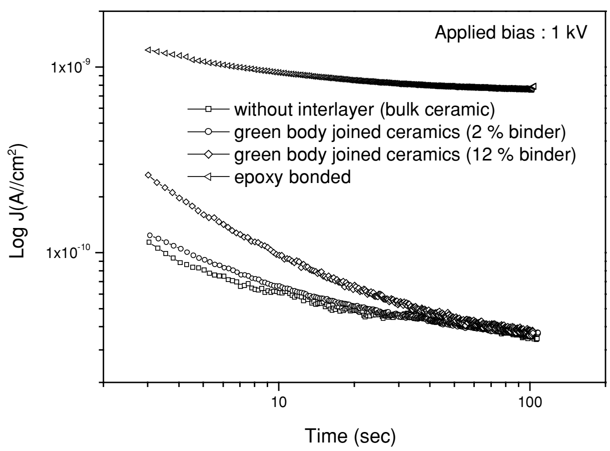

Fig. 3 Time dependent current behavior of type(I) without interlayer (bulk ceramics), type(II) green body joined ceramics with different composition of binders, and with epoxy bonded ceramics

Figure 2 display the voltage dependent leakage current behavior of type (I) bulk ceramics, type (II) green body joined PMN-PZT ceramics with different binder composition, and type (III) epoxy bonded ceramics. As shown in the figure, leakage current levels in green body joined ceramics and bulk ceramics did not changed. However, epoxy bonded structure shows 4 times increased leakage current density at electric field of 1 kV/mm. We ascribe this increased leakage current level to the epoxy material. Since this epoxy has very weak electric field strength, leakage current level was increased at the high electric field.

Figure 3 shows time dependent leakage current behavior of type (I) without interlayer (bulk ceramics), type (II) green body joined ceramics with different 2, 6, 12, 24 wt% binders. type (III) epoxy bonded ceramics. Electric field of 1 kV/mm was applied to the ceramics plates to analyze time dependent electric field behavior. Green body joined ceramics have similar leakage current level with that of bulk ceramics. As shown in the figure leakage current level of epoxy bonded ceramics have almost 5 times higher than those of green body joined ceramics and without interlayer ceramics (bulk ceramics). Since this time dependent leakage current levels were recorded under high electric field of 1 kV/mm for a long time, the leakage current level between the green body joined ceramics and epoxy bonded ceramics was increased compared with Fig. 2 (current-voltage graph). We can say that green body joined ceramics and bulk ceramics has negligible different in electric field strength even in the high electric field.

Impedance data of materials have capacitive and resistive components, when represented in the Nyquist [10] diagram lead to a semicircle. Therefore polycrystalline materials usually represent grain and grain boundary properties leading to semicircle with two RC parallel circuits having different time constants. Figure 4 shows complex impedance plots of without interlayer ceramics (bulk ceramics), green body joined ceramics, and simulated values, respectively. Complex impedance plot of epoxy bonded ceramics did not displayed in this figure, since it have too high impedance value to draw in Fig. 4. The insert shows equivalent circuit for the series connected two parallel connected circuits for bulk and grain boundary, respectively. The insert equivalent circuit was employed to simulate with the measured value. The symbols of □, and ○ represents measured data of bulk ceramic, and green body joined ceramics with 12% binder, respectively. As shown in the figure bulk ceramics and green body joined ceramics can be represented in two semicircles. One RC parallel circuit represents bulk properties and the other RC parallel circuit represents grain boundary properties, respectively. From the simulated line in the figure capacitance of C_1 and C_2 have very small difference and resistance of R_1 and R_2 have 2.7 times differences. It means that bulk and green body

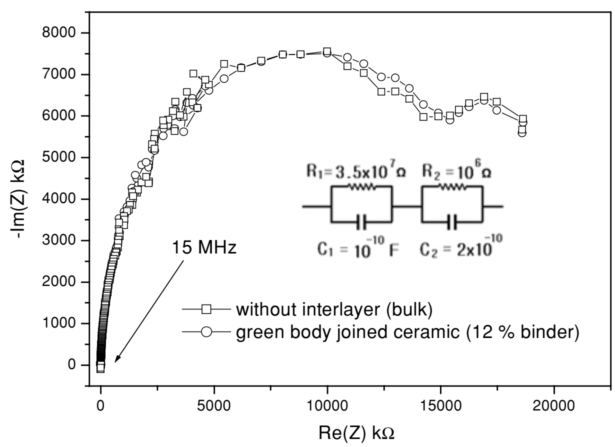

Fig. 4 Complex impedance data of without interlayer ceramic (bulk ceramic), green body joined ceramics, and simulated value with series connected with two parallel circuit. Symbols of □, and ○ represent bulk ceramic, and green body joined ceramic, respectively. The insert circuit is the equivalent circuit for the bulk and grain boundary for the PMN-PZT ceramics

joined ceramics have very similar electrical properties in the impedance data.

4 Conclusion

Green body joined ceramics have very similar electrical properties such as polarization (P) versus electric field (E) characteristic, current (I)—voltage (V) characteristic, and time dependent current characteristics with bulk ceramics, while epoxy bonded ceramics showed totally different electrical properties. Complex impedance plot showed that green body joined ceramics have almost similar grain and grain boundary properties with bulk ceramics. Proposed joining method shows no cracking behavior during the sintering process. The electric properties of green body joined ceramics have improved compared with epoxy bonded ceramics, which mostly used in the commercial bonding materials. Although organic additives with different binder systems are used as joining materials**,** all binder systems of ceramic slurry are available for joining condition.

Acknowledgments This research has been supported by a grant from the Center for Advanced Materials Procesing (CAMP) by the Ministry of Science and Technology and by the Research Grant of Kwangwoon University in 2005.

References

1. H. Yamaguchi, *J. Am. Ceram. Soc*., **82**, 1459 (1999).
2. J.-H. Koh, S.-J. Jeong, M.-S. Ha, and J.-S. Sung, *Ceramics International*, **30**, 1863 (2004).
3. J.-H. Koh, S.-J. Jeong, M.-S. Ha, and J.-S. Sung, *J. Appl. Phys*., **96**, 544 (2004).
4. J.-H. Koh, S.-J. Jeong, M.-S. Ha, and J.-S. Sung, *Sensors and Actuators A*, **112**, 232 (2004).
5. M.L. Santella, *Am. Ceram. Soc. Bull*., **71**, 947 (1992).
6. W.B. Hanson, K.I. Ironside, and J.A. Fernie, *Acta Mate*., **48**, 4673 (2000).
7. D. Janikovic, P. Sebo, P. Duhaj, and P. Svec, *Mater. Sci. Eng*., 569 (2001).
8. D. Travessa, M. Ferrante, and G. Den Ouden, *Mater. Sci. Eng.* A, 287 (2002).
9. T. Besshi, T. Sato, T. Tanaka, and J. Tamatani, *J. Mater. Processing Technol.*, **132**, 169 (2003).
10. S. Lanfredi and A.C.M. Rodrigues, *J. Appl. Phys.*, **86**, 2215 (1999).