Investigation of The Piezoelectric Charge Coefficient d_{33} of Thick-Film Piezoelectric Ceramics By Varying Poling and Repoling Conditions

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Abstract. Piezoelectric ceramics are commonly used in various sensing applications. In this paper, the effect of poling and repoling conditions on thick-film piezoelectric ceramics were investigated. The piezoelectric charge coefficient of the piezoelectric ceramics were measured with varying poling conditions, where the effect of changing poling temperature and electrical field on the d_{33} were analyzed. This was followed by investigating on the effect high applied electrical fields results in repolarization the alignment of the piezoelectric domain in the opposite direction. The temperature and electrical field dependence polarization of the thick-film piezoelectric ceramics were varied near to its Curie temperature between 50°C to 250°C and at a range of electrical field from 20 V (400 kV/mm) up to 200 V (4 MV/mm). It was found that the piezoelectric properties increases with increasing the poling electric field and poling temperature significantly. The maximum values of piezoelectric coefficient were obtained for the piezoelectric ceramics poled at the Curie temperature with high electric fields for 15 min. The aging behavior of the piezoelectric ceramics shows that piezoelectric charge coefficient d_{33} depends on the poling and repoling conditions.

Keywords: poling, repoling, piezoelectric, piezoelectric charge coefficient d_{33}
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INTRODUCTION

The piezoelectric ceramic material converts mechanical energy to electrical energy (direct piezoelectric effect) which makes them useful as sensors. It also converts electrical energy to mechanical energy (indirect piezoelectric effect), which makes them useful as actuators [1], [2]. Hence, the piezoelectric ceramics have been known in electronic devices and extensively used as sensor and actuator applications because of their excellent piezoelectric properties [3], good reproducibility [4], low production cost [5] and it has high potential as smart materials [6]–[8]. Furthermore, in diversify the piezoelectric properties for different kind of purposes, the effect of dielectic characteristics and piezoelectric properties of ceramics material by various conditions have been investigated [2], [9]–[12].The variation in processing parameters including heating, cooling, sintering, fabrication and degradation have also been conducted to find the optimum characteristics of piezoelectric behavior [13]–[17]. However, the effect of the piezoelectric properties via vary poling and repoling process to find of the optimization conditionsfor thick-film piezoelectric ceramic are not well established yet [18], [19].

In order to observe the effect of charge coefficient, mean to disclose these poling variables on thick-film piezoelectric ceramic and approach a possible to optimize for poling and repoling condition process. The poling of thick-film piezoelectric ceramics is intended here to get a net polarization in a certain direction by applying high DC electric fields to the piezoelectric ceramics with temperature (near to its Curie temperature). The nucleation and growth of domains aligned with the applied electric field occurs [20]–[22]. The extent of alignment is dependent on the conditions of poling temperature, applied external electric fields and poling duration, and determines the piezoelectric properties obtained. The poling and repoling condition is required to complete the poling vary with the kinds of piezoelectric ceramics. The control of polarization process is an important parameter to prepare useful and high performance piezoelectric ceramics [23], [24]. A piezoelectric ceramic when subjected to an external electric field undergo change in dimensions because of the displacements of positive and negative charges within the material. Due to this situation, the materials are made up of cations and anions connected by the chemical bonds. When an electric field is applied to the material, the cations get displaced in the direction of the electric field and the anions get displaced in the opposite direction which resulting in net deformation of the material. The piezoelectric

ceramic is a dielectric material because asymmetric displacement of anions and cations. The piezoelectric ceramic materials are noncentrosymmetric and the movement of cations and anions cause the material possessing a centre of symmetry (unit cell) when subjected to an external electric field. The extension and contraction get canceled between neighbouring chemical bonds and net deformation in the material is ideally nil.

Generally, the higher biasing electric field results more complete polarization. In a conventional method, piezoelectric ceramics are poled by applying an appropriate direct-current (DC) electric fields of 2 - 500 kV/mm at elevated temperature around 80^{o}C - 140^{o}C for 5 - 30 minutes [12], [13], [18], [23], [25], [26]. Due to the anharmonicity of the chemical bonds, there will be an electrostrictive effect but it is masked by the more significant asymmetric displacement. The change in dimension is depending on the material which could be very small or quite significant. These materials belong to the class of smart material because they exhibit inherent transducer characteristics [27].

Initially, the piezoelectric ceramic material consists of a large number of ferroelectric domains. Each domain (microscopic region) has a specific polarization direction. Normally, all the domains in the piezoelectric ceramic material are randomly oriented and cause the net polarization of the material is zero in the absence of an electric field (Fig.1 (a)). When the materials are subjected to an external electric field, each atom or molecule in the material possesses an electric dipole moment as the centres of positive and negative charges are separated by a certain distance. The microscopic ferroelectric domains tend to be aligned by applying an external electric field and the domain direction of alignment gets oriented themselves in the direction of the applied external electric field (Fig.1 (b)). As the electric field is increased, almost all the domains get oriented with line up parallel each other in the same direction form a single large domain (Fig.1 (c)). The piezoelectric ceramic material possesses the maximum polarization in this phase. This process is also known as poling. If the piezoelectric ceramic material is maintained at a high temperature near to the Curie temperature while the sufficient electric field is applied, the orientation of the domain alignment is better facilitated and develops a finite polarization [19], [28]–[29].

In addition, the piezoelectric ceramic material can be poled and repoled by the process of generating net remnant polarization in the piezoelectric ceramic material by applying sufficiently high electric field. The chance of damaging the poled piezoelectric materials by breakdown is also increased. Similarly, longer poling duration and higher poling temperature derive better piezoelectric properties. The factors affecting the piezoelectric behavior also may be possibly correlated to the poling and repoling conditions leads to investigation of the piezoelectric charge coefficients (d_{33}) in the thick-film piezoelectric ceramic. In addition, the processing efficiency and operation of an equipment may be difficult to be controlled with respect to the interested to find an economical and effective condition process. All of them are the purposes of the present study.

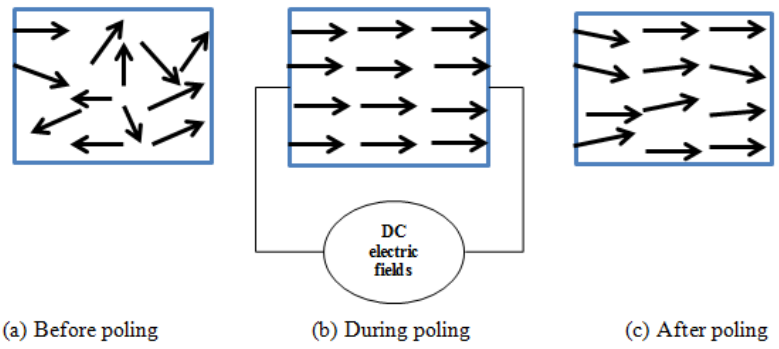

FIGURE 1. Poling of piezoelectric ceramic material : Each arrow represents a ferroelectric domain. (a) Unpoled state – the domains are initially aligned randomly. (b) The high DC electric field is applied to the piezoelectric ceramic material – the domains get oriented in the direction of the electric field. (c) The electric field is removed – most of the domains remain aligned.

EXPERIMENTAL DETAILS

In the present study, the thick-film piezoelectric ceramic is characterized in both poling and repoling process conditions. In the poling process, applied high direct-current (DC) electric field, temperature and time are variable. The thick-film piezoelectric ceramic were treated using the following process. Variations of biasing fields 20 V to 200 V or (400 kV/mm to 4 MV/mm) and poling duration (5 to 25 min) are applied to investigate the piezoelectric properties and find the optimal poling process. To describe the optimal poling conditions, the thick-film of piezoelectric ceramic were poled under different conditions (electric field, temperature and time) and the d_{33} coefficient was monitored as a function of time (25 minutes). Instead of employing specialized poling equipment, the experiment of poling and repoling conditions of thick-film piezoelectric were conducted by a simple method that uses an external high voltage power supply (Matsusada) and hot plate (Fisher Scientific). Since poling at elevated temperature could be done inside the interferometer, the thick-film piezoelectric ceramic were poled on the hot plate, and the piezoelectric properties measurements started when the piezoelectric ceramic is cooled to room temperature after poling was finished. Hence, the advantages of this study thus largely eliminates the costly equipment and time-consuming procedures involving in the conventional treatment and development of sensor systems.

In addition, to demonstrate the piezoelectric properties and measure charge coefficient after the polarization process, characterization on aging rate experiments were performed on the thick-film piezoelectric ceramic by continuous dynamic applied force to the poled samples for 2 hours. The specification of thick-film piezoelectric ceramic dimension is (16.5 mm long x 10.5 mm wide x 1.0 mm thick). The thickness of piezoelectric ceramic layers was approximately 50μm. The piezoelectric layers can be poled to the maximal polarization to normalize the piezoelectric ceramic. Due to the layer structure of thick-film piezoelectric ceramic, the external voltage at the top and bottom electrode of conducting layers generates an electric field that poles the middle piezoelectric layer. The thick-film piezoelectric ceramic was poled simultaneously. The thick-film multilayer structure can be efficiently poled with a high applied external voltage.

On the other hand, the repoling process enhances the net piezoelectric effect in the thick-film piezoelectric ceramic layer, thus increasing the charge coefficient and sensitivity in sensor applications. The piezoelectric ceramic materials have an initial polarization but the net polarization is variable and minor. The piezoceramic samples are first heated to the temperature needed (50^{o}C to 250^{o}C) on a hot plate (Fig.2 (a)) for 2 min to eliminate the initial polarization for better polarization alignment. To pole and repole the piezoelectric thick film, the variation of biasing fields 400 kV/mm to 4 MV/mm were then applied to the samples via the soldering tags and conducting layers for the various poling duration (5 to 25 minutes) while holding them at the temperature needed (near to its Curie temperature) (Fig.2 (b)). Lastly, the piezoelectric ceramics were removed from the hot plate while the high applied DC electric fields are maintained connected to the electrodes of the piezoelectric ceramics and allowed to cool to room temperature (Fig.2 (c)). The electric field is then unconnected after a few minutes when the piezoceramic samples cool to room temperature and remains in the maximum polarization state with most of the domains oriented in the same direction.

Generally, higher polarization of the piezoelectric ceramic layer results in a greater charge coefficient for the piezoelectric devices. In addition, by using this polarization process, the piezoelectric ceramic can be treated to saturation which is relatively high polarization level and exhibits good piezoelectric behaviors.

The measurement procedure was as follows. The poled piezoelectric ceramic were tested using the mechanical testing machine. The piezoelectric charge coefficients (d_{33}) were characterized using a quasi-static d_{33} meter (ZJ-6B, Institute of Acoustics, Chinese Academy of Sciences, China). The poled samples of piezoelectric ceramic were observed for 2 hours and the piezoelectric charge coefficients (d_{33}) were measured again to study the aging behavior of thick-film piezoelectric ceramics.

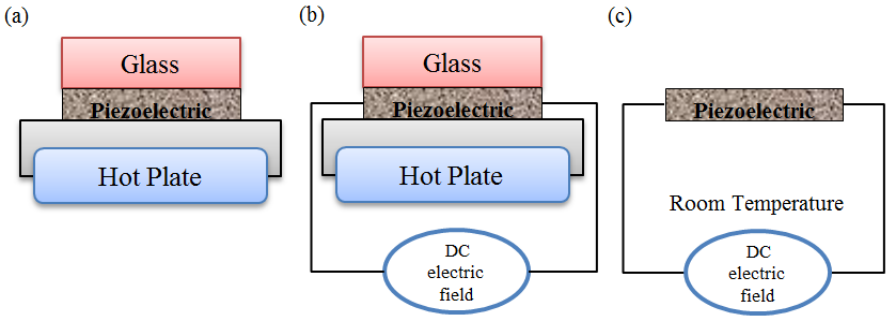

FIGURE 2. Illustration of the poling and repoling conditions for the thick-film piezoelectric ceramic:(a) Piezoelectric ceramic material heated at a certain temperature. (b) A DC electric field is applied to the piezoelectric ceramic with the certain temperature needed. (c) A DC electric field is maintained to the piezoelectric ceramic during cooling to the room temperature.

RESULTS AND DISCUSSION

Fig. 3 presents the variation in the piezoelectric charge coefficient,d_{33} of the thick-film piezoelectric ceramic. The piezoelectric ceramic were characterized using a quasi-static d_{33} meter with poling time ranging from 0 minutes to 25 minutes. The effect of piezoelectric properties of the thick-film piezoelectric ceramic on the poling and repoling condition were observed. The samples were poled were at 200 V (4MV/mm) with temperature 200°C for both poling (P_{ori}) and repoling (P_{new}) condition. It was found that the piezoelectric charge coefficient of ceramic increases with increasing poling duration until it reaches toward the saturation state (dotted line). In the beginning, as can be seen the d_{33} value was 0 pC/N due to domain are initially aligned randomly for poling condition (P_{ori}) compared to repoling condition (P_{new}) where the d_{33} value was 12.2 pC/N due to the initial polarization. The piezoelectric charge coefficient, d_{33} (pC/N) of thick-film piezoelectric ceramic can be efficiently raised and normalized at 15 minutes via both conditions.

However, the piezoelectric charge coefficient of thick-film piezoelectric ceramic were slowly increasing after 15 minutes to 25 minutes for the poling condition (P_{ori}) process and the mean piezoelectric charge coefficient, d_{33} value was 24.5 pC/N at 25 minutes, which is in saturation state. Meanwhile, the charge coefficient of piezoelectric ceramic slightly increases after 15 minutes to 25 minutes for the repoling condition (P_{new}) process and the mean piezoelectric charge coefficient, d_{33} value was 36.2 pC/N at 25 minutes, which is significantly higher than poling condition (P_{ori}). The mean piezoelectric charge coefficient, d_{33} value were 4.4% difference and 2.4% difference from the saturation state for both poling (P_{ori}) and repoling condition (P_{new}) respectively at 15 minutes. Thus, the poling duration also affects the piezoelectric behavior which requires a longer time to treat the ferroelectric domains get oriented in the direction of the electric fields [30].

In order to investigate the effect of thick-film piezoelectric ceramic in the poling condition, the samples were poled at various poling electric fields and various poling temperature. In the beginning, the samples were poled with various poling temperature, but a constant of applied electric fields at 4MV/mm for 15 min poling duration. The increasing of poling temperature is an effective way to obtain high piezoelectric coefficient in the piezoelectric ceramic material. Fig.4 reveals the piezoelectric charge coefficient, d_{33} were measured as a function of temperature. The temperature dependence of the piezoelectric response as can be observed, which leads to significantly increased the piezoelectric charge coefficient,d_{33}. The experiments were carried out by starting at 50 °C with ranged to 250°C. The results demonstrated the poled piezoelectric response for 0 min (at the beginning of measurement) and 120 min respectively. At the variation of 0 min, the piezoelectric charge coefficient of thick-film gradually increasing as the temperature increases until the arrives to the peak value of the piezoelectric charge coefficient, d_{33} was 36.2 pC/N at 200°C. The piezoelectric charge coefficient of thick-film was also decreased dramatically to 24.4 PC/N at 220°C and then increase again to 25.3 pC/N at 250°C. Meanwhile, at the variation of 120 minutes, the results also shown the same pattern as the result at variation of 0 min but it reaches to its peak value of the piezoelectric charge coefficient, d_{33} was 25 pC/N at 180°C. The piezoelectric charge coefficient is slowly decreases to 15.6 pC/N at 220°C and then rise again at 250°C with the d_{33} value 18.3 pC/N.

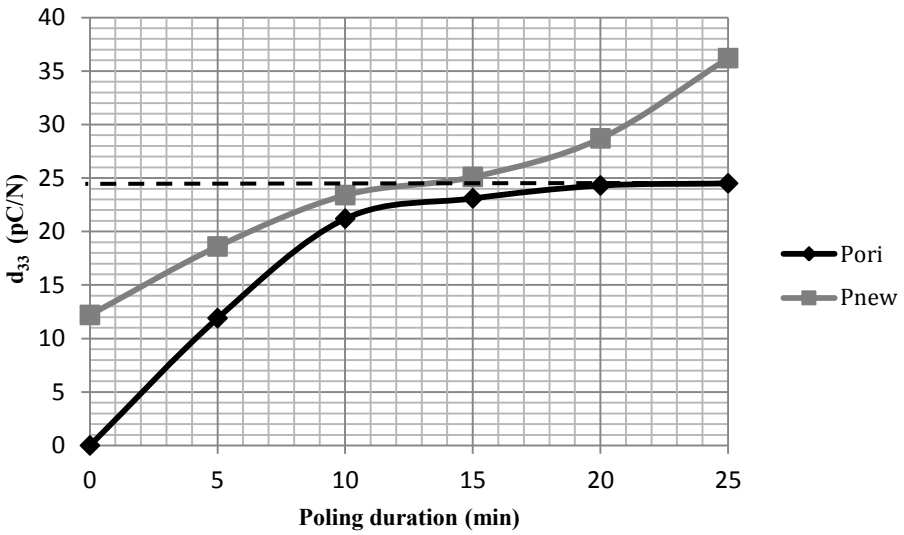

FIGURE 3. The variation in the piezoelectric charge coefficient,d_{33} of the thick-film piezoelectric ceramic of the poling (P_{ori}) and repoling (P_{new}) conditions with various poling durations.

Compared to the both results, this feature indicates that the piezoelectric ceramic easily polarized to saturation state with applied high temperature (near to the Curie temperature). On the other hand, the fact that some of ferroelectric domains walls were difficult to align because of the thicker piezoelectric ceramic layer and low temperature applied [31], [32]. In addition, the mobility of ferroelectric domain is greatly increase which allowing their oriented to better alignment in the direction at high temperature. However, too high temperature make the decreased of sensitivity of piezoelectric and the extension a little bit of ferroelectric domains alignment due to the movement of dipole [33]. It can be shown that the thick-film piezoelectric ceramic was significantly affected by the temperature. In this experiment, the optimization of the temperature can be observed was around 200°C for the thick-film piezoelectric ceramic.

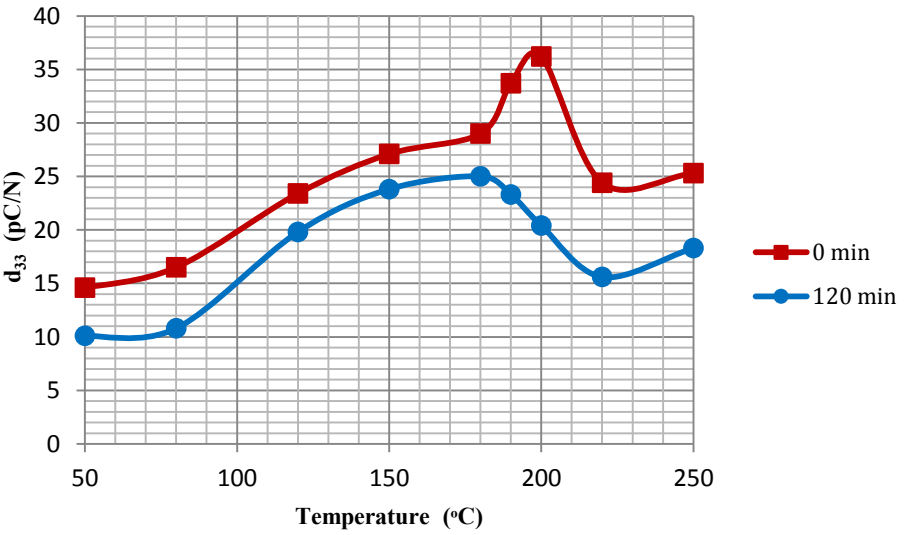

FIGURE 4. The variation in the piezoelectric charge coefficient,d_{33} of the thick-film piezoelectric ceramic at 0 min and 120 min conditions with various temperature, respectively.

To optimize the both poling conditions, the samples were polarized with the different poling electric field (MV/mm), but a fixed poling temperature at 200°C for 15 min poling duration. The applied electric field dependence piezoelectric properties as can be shown. Fig.5 represents the maximum piezoelectric charge coefficient,d_{33} (pC/N) of thick-film piezoelectric ceramic vary with the applied electric field. The experiments were carried out by starting with ranged the electric fields from 0 MV/mm to 4 MV/mm. The piezoelectric properties was decreases when the applied electric decreases due to the inadequate switching of the ferroelectric domain. In fact, it's also because of the thicker of piezoelectric layers make the ferroelectric domain difficult to align in the direction [34].

Indirectly, the results demonstrated that as the electric field increases, it also significantly increased the piezoelectric charge coefficient. So, the higher electric field, the more oriented ferroelectric domains and higher piezoelectric properties. However, excessive the external electric applied to the thick-film piezoelectric leads to overpole the ceramic material. This phenomenon can cause the physical flaws to piezoelectric ceramic close to the dielectric breakdown of the material [27]. These results demonstrated that the thick-film piezoelectric ceramic can be poled and repoled depends on the polarization temperature, sufficiently applied electric field and poling time duration. So, the more oriented ferroelectric domains, the higher piezoelectric properties.

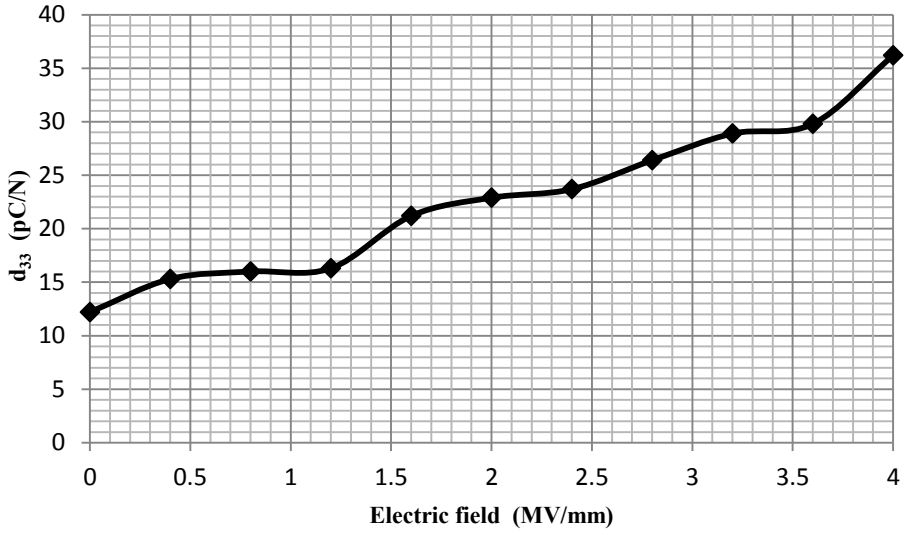

FIGURE 5. The maximum piezoelectric charge coefficient,d_{33} (pC/N) of thick-film piezoelectric ceramic vary with the applied electric field.

For the aging rate investigation, the effect of piezoelectric properties of the various poling temperature were evaluated. The piezoelectric ceramic were treated in a constant electric field 4 MV/mm with various temperature ranging 50°C to 250°C and then continuously measure for 2 hours. Fig. 6 reports the response of aging behavior on the thick-film piezoelectric ceramic in a constant applied electric field of 4 MV/mm with various poling temperature for 2 hours continuously applied dynamic force of 1 N. The highest charge coefficient of thick-film piezoelectric ceramic was 36.2 pC/N can be observed at 200°C of temperature in the beginning of time with further increases the continuous time for 2 hours, the piezoelectric coefficient was 20.4 pC/N which is exponentially decreasing after 1 hours and then gradually decreases over the time in another 1 hours.

It is seen that, at 250°C and 220°C the d_{33} value were 25.3 pC/N and 24.4 pC/N for the beginning, respectively. It is not a high piezoelectric charge coefficient, even its measure at high temperature such as 250°C and 220°C because of loss of polarization due to too high temperature. For temperature 180°C and 150°C, the piezoelectric response shows that the piezoelectric properties was slowly decreased almost the same pattern as the time increases. For the temperature 80°C and 50°C, the piezoelectric coefficient were 16.8 pC/N and 14.6 pC/N at the beginning, respectively, and as the time increases to 2 hours, the results response is exponentially decreasing with d_{33} value 10.8 pC/N and 10.1 pC/N, respectively. These response results demonstrated that the piezoelectric properties decayed over the time.

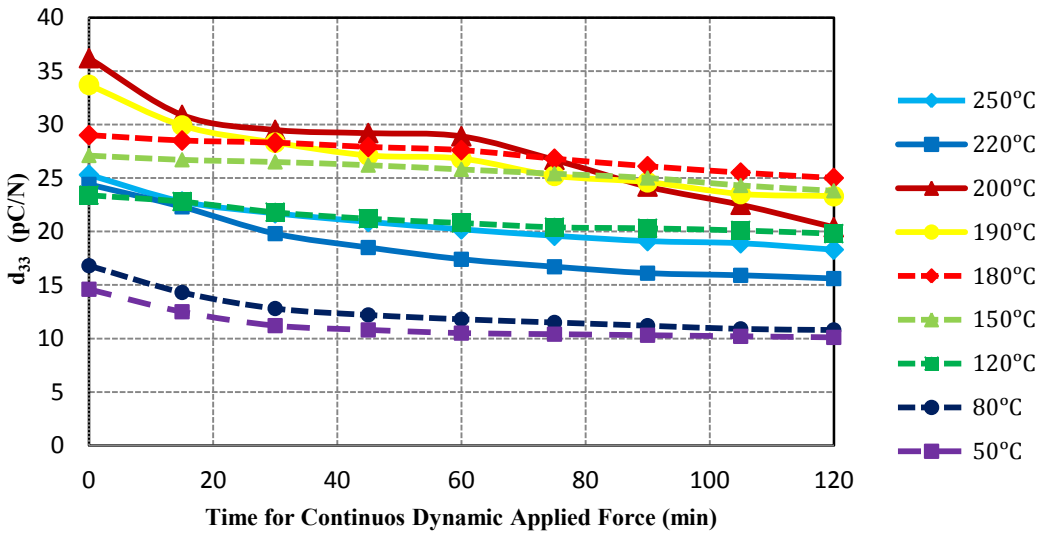

FIGURE 6. The variation in response of aging behavior on the thick-film piezoelectric ceramic with various poling temperature in a constant applied electric field of 200 V .

Finally, the aging behavior of piezoelectric properties of various the poling electric fields was evaluated. The piezoelectric ceramic were treated at a constant temperature of 200°C with various poling electric ranging from 0 MV/mm to 4 MV/mm and continuously measure for 2 hours. It is seen that, from the beginning the piezoelectric charge constant is higher and gradually reduced over time. Fig. 7 shows the response of the aging behavior of thick-film piezoelectric ceramic with various poling electric fields in a constant temperature of 200°C for 2 hours continuously applied dynamic force of 1 N.

These aging response results also play the same role as aging investigation of the various temperature whereby demonstrated that the piezoelectric properties decayed over the time. The highest charge coefficient of thick-film piezoelectric ceramic was 36.2 pC/N can be observed at the beginning of time for 4 MV/mm with increases the time to 2 hours, the piezoelectric coefficient was exponentially decreased to 20.4 PC/N. For 3.6 MV/mm, the piezoelectric charge was rapidly decreasing for earlier 30 minutes 28.9 pC/N to 16.4 pC/N and then with increasing to time it slowly decreases to 13.5 pC/N. This condition also happens to this variable from 3.2 MV/mm to 1.6 MV/mm. For variable 1.2 MV/mm and 0.8 MV/mm, the piezoelectric properties were slightly exponentially decreases with increasing time. However, it can be observed that when continuously applied dynamic force of 1 N for 2 hours, the properties were almost remained at the same level, but actually the piezoelectric constant very slowly decreased from 12.2 pC/N to 10.7 pC/N without applying electric field 0 MV/mm because of the initial polarization of thick-film piezoelectric ceramic.

CONCLUSIONS

The piezoelectric properties of thick-film piezoelectric ceramics were successfully investigated by varied poling and repoling conditions. A simple method was used to enhance the piezoelectric performance of the thick-film piezoelectric ceramic and largely eliminates the time-consuming procedures. The poling electric fields, the poling temperature, poling duration have shown great influence in the performance of the piezoelectric ceramic material. The direct piezoelectric charge coefficient d_{33} was significantly increased as the applied electric field is increased toward the saturation point. The charge piezoelectric coefficient also increases after poling under high temperature was observed. It was observed that the slightly over pole effect in thick-film piezoelectric ceramic at high temperature. The piezoelectric charge coefficient exhibits a rapid increase for poling fields from 60 V (1.2 MV/mm) to 200 V (4 MV/mm). The piezoelectric constant showed a maximum value of d_{33} ~ 36.2 pC/N. Hence, the optimum poling and repoling condition for the thick-film piezoelectric ceramics is subjected to applied electric field and poling temperature are around 200 V (4 MV/mm) and 200°C, respectively, with 15 min of poling duration. The aging behavior of thick-film piezoelectric ceramic were successfully demonstrated via varying electric fields and

varied temperature. It was found that the mean piezoelectric properties of thick-film piezoelectric ceramics was decreased upon poling which is corresponding to the significant aging response. The effect can be useful for poling and repoling conditions piezoelectric ceramic based thick-film piezoelectric and this study suggest that a significant increment of polarization plays an importance role in high performance piezoelectric properties.

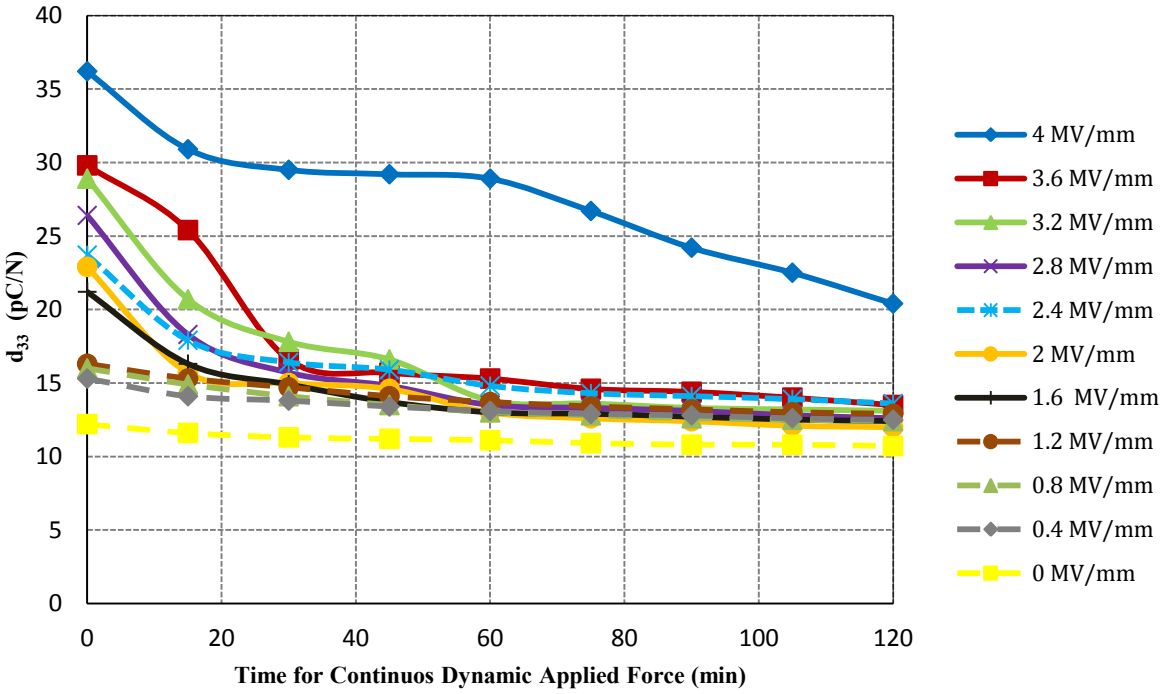

FIGURE 7. The variation in response of aging behavior on the thick-film piezoelectric ceramic with various applied electric fields for 2 hours in a constant temperature of 200°C.

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