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A Brief Review of Some Challenging Issues in Textured Piezoceramics via Templated Grain Growth Method

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Abstract

It is well known that polycrystalline ceramics fabricated via the templated grain growth method along a desired crystallographic direction, generally along [001], exhibits enhanced piezoelectric response. Generally, the piezoelectric properties of textured ceramics depend on the degree of texture, as piezoelectric properties peak in single crystals. Therefore, understanding the relationship between the degree of texture and piezoelectric properties is fundamental. Here, we present state-of-the-art textured piezoceramics by focusing on critical issues such as the quality of templates used for texturing and proper evaluation of the degree of texture analysis. The relationship between the degree of texture and its impact on the properties of textured materials is exclusively defined by the Lotgering factor (L.F.) calculated from the X-ray diffraction profiles. Additionally, we show that L.F. is not a suitable indicator of the degree of texture, contrary to previous interpretations. This statement was further supported by the fact that the true degree of texture can be better quantified by the multiples of random distribution. This argument was justified by comparing the quantitative values of the degree of texture obtained from both methods to those of the piezoelectric charge coefficient of textured and random ceramics.

Keywords: Piezoceramics, Templated grain growth, Lotgering factor, Multiples of random distribution

1. INTRODUCTION

Piezoelectric materials can convert mechanical energy into electrical energy, making them one of the most in-demand materials in state-of-the-art technological technologies [1, 2], such as robotics, consumer electronics, ultrasound transducers, sensors, and actuators, etc. [3, 4]. To enhance the applicability of piezoceramics in such applications, studies have been conducted on enhancing their performance and lowering manufacturing costs. In general, piezoceramics exhibit significantly lower performance than their single-crystal counterparts, owing to the random crystallographic orientation of the grains [5]. Despite their excellent performance, single crystals are expensive and difficult to manufacture, due to compositional fluctuations during manufacture. The shapes and sizes may also be limited due to the low mechanical strength [6], and these shortcomings limit their applications.

Over the years, intensive investigations have been conducted to overcome the limitations of piezoelectric single crystals and ceramics. This led to the discovery of textured ceramics, in which the grains are oriented along a preferred crystallographic direction, especially via the templated grain growth method (TGG), realized by doctor-blade-based tape-casting processes [7]. TGG is considered the most effective method for inducing grain orientation, as it can produce higher levels of texture with better alignment in the intended crystallographic direction among the grains, compared to traditional methods such as hot forging and hot pressing of anisotropically shaped particles [7]. Textured ceramics fabricated using the TGG method have a single crystal-like structure, except for the presence of the grain boundary between oriented grains and phase boundary between textured grains and embedded templates, as schematically illustrated in Fig. 1 [7]. This allows access to single crystal-like properties with better mechanical strength, and the possibility of fabricating a wide range of chemical compositions of various shapes and sizes at a lower cost than that for single crystals [10]. To this end, several piezoceramics have been textured with the aim of leveraging texturing, as reflected in the number of publications by year (Fig. 2) [4].

Although single crystals exhibit the piezoelectric properties, the properties of textured ceramics depend on the degree of texturing,

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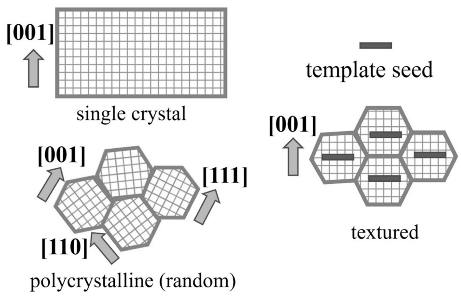

Fig. 1. A schematic of crystal-bearing in the constituent grains of polycrystalline and textured materials in comparison with a single crystal.

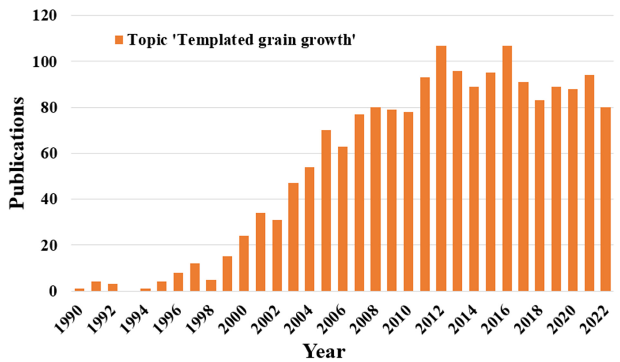

Fig. 2. Publication year versus the number of publications according to Web of ScienceTM (https://apps.webofknowledge.com).

which is strongly affected by the template morphology. Moreover, it is required that the templates are chemically stable, and have suitable crystallographic matching with the target materials [11-15]. Therefore, several studies have been conducted to optimize the size, size distribution, shape, and aspect ratio for better alignment during tape casting processes [11-15]. However, even in highly textured ceramics, the piezoelectric performance is significantly lower than that of single crystals of the same composition. This is an issue of significant research interest, as reflected in the statistics on the number of publications by year (Fig. 2). This may be indicative of the inability of textured ceramics to replace expensive single crystals, which has led to the search for other underlying factors conventionally not considered in texturing. One of such factors is the structural quality of the template used for texturing, because the structure of the textured grains is highly affected by it. More importantly, quantitative determination of the degree of texture should be considered to understand its fundamental relationship with the properties. In most textured piezoceramics, the degree of texture is determined by calculating the Lotgering factor (L.F.) from the X-ray diffraction (XRD) profiles of both random and textured ceramics [7]. However, the reliability of this method for determining the degree of texture must be cross-checked with alternative methods. In this review, we present state-of-the-art textured piezoceramics, focusing on the structural quality of templates for a better degree of texture. More importantly, methods for determining the degree of texture and their relationship with the changing properties are also discussed.

2. ISSUES ASSOCIATED WITH THE TEMPLATES USED FOR TEXTURING

The selection of an optimal template crystal for texturing a specific piezoelectric ceramic requires careful consideration, as the crystal structure of the template must be the same or very similar to that of the ceramic to achieve successful texturing. This is because the template acts as a model for the growth of the ceramic crystals, and if the crystal structures are not matched, the texturing process will not be effective. Additionally, lattice mismatch, which refers to the difference in the crystal lattice spacings between the template and the ceramic, should also be considered as it can have a significant impact on the success of texturing [7]. More importantly, the template needs to be sufficiently stable in the matrix system to neither diffuse nor cause compositional changes in the target material [7]. These requirements limit the window of template selection for texturing certain piezoceramics. Kwon *et al.* [8] reported that when texturing PMN-0.325PT ceramics using $SrTiO_3$ (ST) or $BaTiO_3$ (BT) plate-like crystals, the ST template was found to be chemically less stable than BT in a matrix system during sintering, resulting in a compositional change in the target material, which resulted in a shift in the Curie temperature (T_C), as shown in Fig. 3.

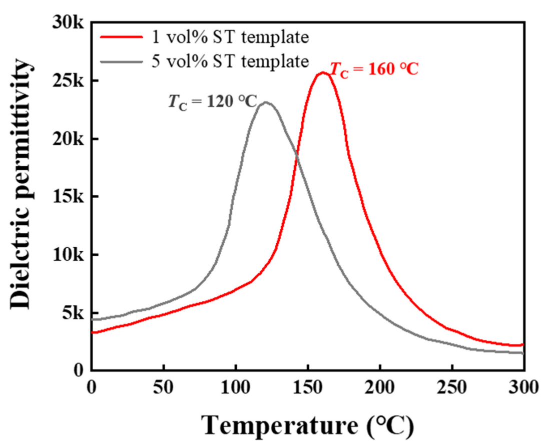

Fig. 3. Temperature-dependent dielectric permittivity of textured PMN-0.325PT ceramics containing 1 and 5 vol% of ST template, as in the work of Kwon *et al.* The permittivity data were retrieved from Ref. [8].

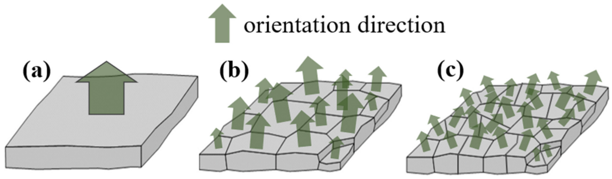

Fig. 4. Schematic of high-quality template, (single crystal) (a), moderate quality template (highly oriented polycrystalline) (b), low-quality template (polycrystalline having relatively poor orientation) (c).

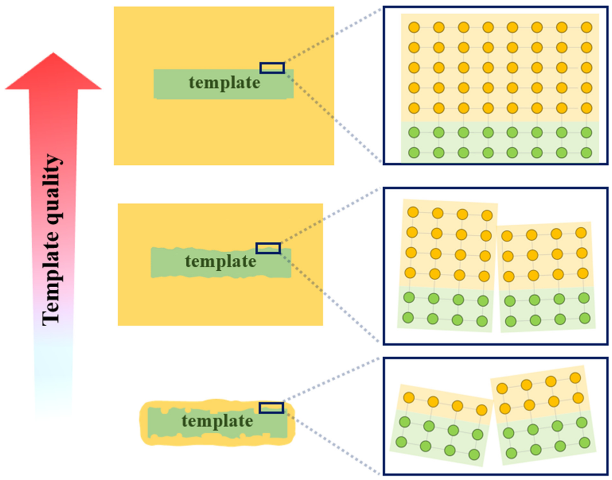

Fig. 5. Schematics manifesting the effect of template quality on the microstructure of TGG.

BT templates have also been used to obtain highly textured PMN-based material systems because of their suitable crystallographic matching and chemical stability during sintering [13,17-20]. Although these are the key parameters for selecting a good template, an additional requirement is that the template should be a pure single crystal, such that the textured grains assume a preferentially well-oriented structure [21]. However, the template quality has not been addressed adequately in literature, which may affect the quality of texturing, and consequently degrade the performance. Poterala *et al*. [21] reported that the BT template prepared by molten salt synthesis followed by topochemical conversion can form either a single crystal or polycrystalline structure, as shown schematically in Fig. 4.

According to Poterala *et al*. [21], the formation of a polycrystalline structure is associated with the formation of multiple misaligned nucleation sites during the topochemical conversion process, and a single crystal template can be obtained by controlling the synthesis temperature and time. Fig. 5 illustrates how the use of templates of different qualities can deteriorate structural quality during TGG. Therefore, in addition to template chemical stability, crystallographic match, and morphology, critical attention to the structural quality of the template is necessary.

3. ISSUES ASSOCIATED WITH THE DEGREE OF TEXTURE ANALYSIS

The higher the degree of texture, the better the properties of the material [9]. This means that the accuracy of determining the degree of texture is crucial for understanding its impact on properties. In almost all textured piezoceramics, the degree of texture along the (001) orientation was reported in terms of the calculated L.F. value. For samples textured along the (001) direction, the corresponding L.F. can be determined using Eq. (1), which gives a number in the range of 0 to 1 based on the XRD intensity of both random and textured ceramics. [7, 22]:

$$\text{L.F.} = (P_{(00l)} - P_o)/(1 - P_o) \tag{1}$$

$$P_{(00l)} = \Sigma I_{(00l)}/\Sigma I_{(hkl)} \tag{2}$$

$$P_o = \Sigma I_{o(00l)}/\Sigma I_{o(hkl)} \tag{3}$$

where $P_{(00l)}$ and P_0 are the sum of the XRD peak intensities of all the (00*l*) peaks divided by all the (*hkl*) peaks of the textured ceramics, as shown in Eq. (2), and random ceramics, as shown in Eq. (3), respectively. These equations have been widely used to determine the degree of texture of textured ceramics. However, in some cases, the obtained L.F. value is not consistent with the enhancement of properties.

For example, Sabolsky *et al*. [23] reported that textured PMN–0.325PT ceramics with an L.F. of 0.9 showed d_{33} of 1150 pC/N, which is higher than that of random ceramics of the same composition by a factor of 1.95. Similarly, Yan *et al*. [24] reported that textured ceramics of the same composition with L.F. of 0.9 exhibited an increase in d_{33} by a factor of only 1.21, as shown in Fig. 6. This inconsistency indicates that L.F. can be used as a qualitative reference for grain orientation measurement, but cannot be used as a quantitative parameter [25].

The degree of texture can alternatively be obtained from the integrated intensity of specific XRD peaks. Among such methods multiple random distribution (MRD) analysis [25, 26, 28-32] presents the degree of texture for tetragonal symmetry using the MRD value calculated using Eq. 4:

$$f_{002}(mrd) = 3\frac{I_{002}/I^R_{002}}{I_{002}/I^R_{002}2(I_{200}/I^R_{200})} \tag{4}$$

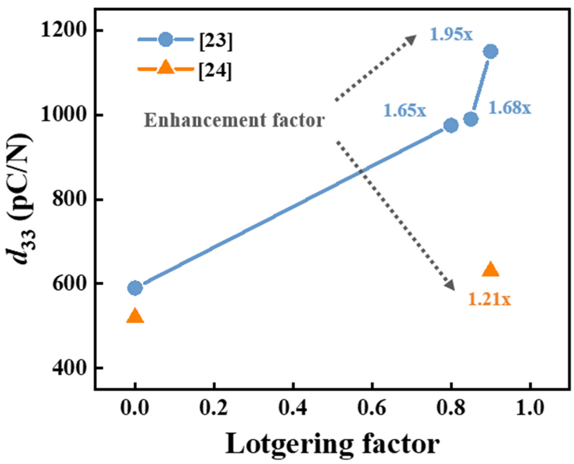

Fig. 6. Variation of d_{33} as a function of the Lotgering factor. The data were retrieved from Refs. [23, 24].

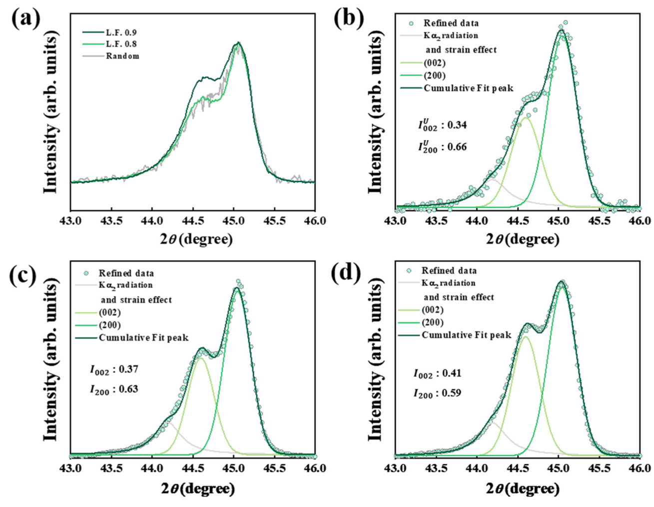

Fig. 7. XRD pattern of the random and textured ceramics (L.F. 0.8 and L.F. 0.9) (a), Lorentz fit peak curve of random (b), textured (L.F. 0.8) (c), texture (L.F. 0.9) (d) Mn-doped PMN-0.29PT ceramics.

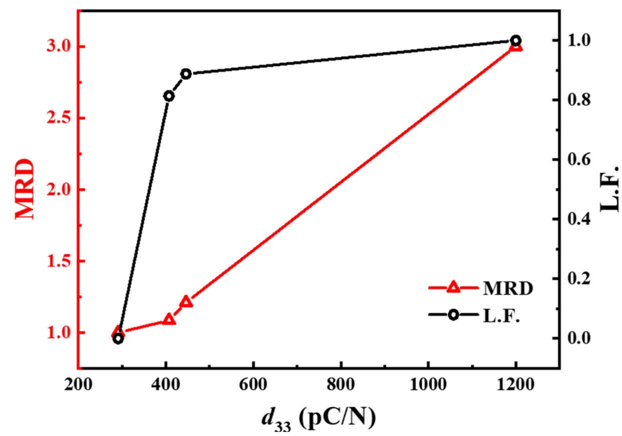

Fig. 8. Comparison of the degrees of texture determined using MRD and L.F. calculation as a function of the piezoelectric coefficients for the random and textured Mn-doped PMN-0.29PT ceramics in reference to a single crystal of approximately the same composition.

Where I^R_{002} and I^R_{200} denote the integrated normalized intensity for random ceramics, whereas, I_{002} and I_{200} are for the textured ceramics [25, 26]. Using the MRD equation, the degree of texture in random and textured Mn-doped PMN-0.29PT ceramics was calculated. Integrated intensities of diffraction peaks in randomly oriented and textured samples were obtained from XRD analysis following Ref. [33]. The Gaussian peak fitting module provided by the commercial software ORIGIN Pro 2020 (Originlab, Inc.) was used to deconvolute $(002)_{pc}$ and $(200)_{pc}$ (the subscript 'pc' denotes the pseudocubic indices) peaks. Due to the limited data points, Cu K_{α_2} radiations were not removed. Instead, a Lorentzian background peak was added to be attributed to the contribution from the Cu K_{α_2} radiations and the presence of a small portion of rhombohedral-originated intensity, as shown in Fig. 7. The obtained integrated intensity of $(002)_{pc}$ and (200) from both random and textured Mn-doped PMN-0.29PT ceramics were used for MRD calculation.

For a proper comparison between MRD and L.F., we measured the piezoelectric charge coefficient d_{33} for four sets of samples of approximately the same composition, i.e., random ceramic, textured ceramic with an L.F. of 0.8 and 0.9, and a single crystal following the standard measurement procedure [34]. The d_{33} of a random ceramic was measured to be 291 pC/N, the L.F. of which was zero. The d_{33} value of the sample with the L.F. of 0.8 and 0.9 was obtained to be 407 pC/N, and 446 pC/N, respectively. It is known that even though ceramics are perfectly textured to have an L.F. of 1.0 or an MRD of 3.0, their d_{33} values are not comparable to with those of the single crystals, owing to the presence of the grain and phase boundaries, as described in the introduction. Nevertheless, we considered a single crystal of the same composition to estimate the d_{33} value for L.F. of 1.0 (MRD of 3.0) as an approximation.

To determine the correlation between the quantitative parameters for the degree of texture and d_{33}, we plotted both L.F. and MRD as a function of d_{33} for the random (L.F. 0, MRD 1), textured (L.F. 0.8, 0.9, MRD 1.11, 1.17) Mn-doped PMN-0.29PT ceramics, and single crystal (d_{33} 1200 pC/N), as presented in Fig. 8. It is evident that L.F. obviously overestimates the degree of texture, while MRD underestimates it. Assuming that the ceramics are perfectly textured at an L.F. of 1.0 (MRD of 3.0), the expected d_{33} value is ~450 pC/N, which is significantly small. IT should be noted that the reported d_{33}

value of PMN-PT ceramics with an L.F. of >0.9 easily surpasses 600 pC/N [35]. By contrast, the estimated d_{33} value from the MRD analysis was ~2150 pC/N, which is even higher than that of a single crystal. It is not certain if the d_{33} value estimated by the MRD analysis is completely unrealistic in that the d_{33} value of single crystals differs significantly from manufacturer to manufacturer. More importantly, the current analysis can be erroneous, because the data used for this analysis are not sufficient to guarantee accuracy. However, it is worth highlighting the fact that the degree of texture estimated by the MRD analyses shows a reasonable correlation with the experimentally measured d_{33} values in comparison with that by the L.F. estimation. There is further scope for developing a method to better estimate the degree of texture, and an MRD-based analysis could be a good starting point for this purpose.

4. SUMMARY AND PERSPECTIVES

Although there are limitations and challenges to overcome, TGG remains one of the most effective approaches for improving the properties of polycrystalline ceramics. Since the properties of textured ceramics are affected by the degree of texture, which mainly depends on the template used, special attention needs to be paid to the template quality in addition to its stability, morphology, and crystallographic match. More importantly, the degree of texture must be carefully examined to understand its fundamental relationship with the properties. L.F. has been used extensively because it is easy and convenient for the determination of the degree of texture. However, as discussed in this brief review, L.F. lacks accuracy. Therefore, to avoid an incorrect interpretation of the fundamental relationship between the degree of texture and these properties, it is strongly recommended not to rely on the L.F. As an alternative, using MRD may be a better approach; however, this does not imply that MRD is the most accurate solution. The calculated values show a relatively consistent relationship with the piezoelectric charge coefficient of textured ceramics. To better visualize the degree of texture, electron backscatter diffraction mapping analysis can provide color impingement crystallographic orientation, serving as an alternative [36]. In addition, since laboratory XRD has poor bulk sample penetration capability, either high-energy X-rays or neutron diffraction could be useful, depending on the type of material [23]. We hope that this brief review will provide fundamental insights to further improve future work on texturing.

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