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STOCHASTIC OPTIMIZATION OF ELECTROMECHANICAL COUPLING IN FERROELECTRIC MATERIALS

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Abstract. Polycrystalline ferroelectrics differ significantly from single crystals because of the presence of variously oriented grains or crystallites. The orientation of ferroelectric crystals plays a critical role in the anisotropy of their piezoelectric properties. The set of combination of variables, known as solution space, which dictates the orientation distribution of grains is unlimited. Thus a stochastic optimization combined with homogenization is employed for the identification of the optimal granular configuration of the ferroelectric ceramic microstructure with optimum electromechanical coupling suitable for applications such as transducers and actuators. The effective macroscopic electromechanical coupling *k* are calculated at every iteration using the mathematical homogenization method. The configuration of crystallites at each step is chosen by the optimization algorithm. The single crystal ferroelectrics are also investigated in an effort to compare the performance characteristics with that of polycrystals. A configuration constraining the orientation distribution of the c-axis (polar axis) of the crystallites is identified. This grain configuration would optimize the figures of merit such as d_h and k_h of the ferroelectric material.

1 INTRODUCTION

Crystalline solids usually exist as polycrystals comprising a large number of individual micro-sized crystallites or grains. The relative position and orientation of the individual crystallites in a polycrystalline substance are usually entirely random. For this reason, when such a substance is considered on a scale large in comparison to the dimensions of the crystallites, it is isotropic. However, this treatment is in contrast to their true molecular anisotropy which takes effect in the individual crystallites. In this paper, we have modeled a polycrystalline ferroelectric material taking into account of the anisotropy at the crystallographic scale. Ferroelectric (FE) crystals undergo asymmetric displacement resulting in a small change in crystal dimension when coaxed by an external electric field. Such electric-field-induced strain called piezoelectricity has extensive applications such as in sensors and actuators and offers great potential for next generation high density storage devices like nonvolatile random access memories (Uchino, 2000; Scott, 2007). It is demonstrated both experimentally (Park and Shrout, 1997; Wada et al., 1999; Wada et al., 2005) and theoretically (Fu and Cohen, 2000; Jayachandran et al., 2008) that a number of ferroelectrics including $BaTiO_3$ (BTO) exhibits an enhanced piezoelectric strain when poled along a nonpolar axis. Polycrystals (ceramics) have engineering advantage over single crystals in technical applications owing to the ease in manufacture and in compositional modifications (Scott, 2007; Damjanovic, 1998). Polycrystal is an aggregate of variously oriented crystallites or grains. The resultant macroscopic polarization in a polycrystal is zero because the polarization directions are randomly oriented, and the material is not macroscopically piezoelectric. If the isotropic non-piezoelectric polycrystal is poled, that is coaxed with a large electric field at high temperature, the directions of polarizations in many of the grains are permanently realigned resulting in macroscopic spontaneous polarization and hence in piezoelectricity (Alguero et al., 2005).

As-grown polycrystalline FE is an aggregate of single crystalline grains with randomly oriented (spontaneous) polarizations (Lines and Glass, 1977). The spatial configuration of crystallographic grains and their orientation distribution (texture) impact the piezoelectricity exhibited by conventional as well as new generation FEs (Scott, 2007). The randomness in polarization-vector orientation renders the resultant piezoelectricity of the material to be marginal or zero. Albeit the resultant polarization is zero for as-grown polycrystal, an overall piezoelectricity can be enabled by the application of an external electric field, called poling field, though all the grains can never align perfectly (Lines and Glass, 1977).

The aggregate texture of an unpoled (as grown) polycrystal would have a uniform random distribution (Brosnan et al., 2006). With the strength of the poling field increases, the nature of the grain orientation distribution becomes Gaussian (normal) (Uetsuji et al., 2004). Nevertheless, the spatial and orientational randomness of grains can be judiciously employed in the design of FE polycrystals (ceramics) with tailor-made configurations. Recently it is shown that overall piezoelectricity would be enhanced by the introduction of either [110]- or [111]-oriented grains into a random $BaTiO_3$ polycrystal (Wada et al., 2007). Despite intense research in ferroelectrics, the solution of optimal grain configuration of the polycrystalline FE is hardly resolved. This is mainly due to the vast number of possible configurations available at hand albeit the FE ceramics are easy to manufacture. This paper is concerned with optimally designing the performance characteristics of the piezoelectric component in a hydroacoustic application such as a hydrophone. We would identify an optimum configuration

of grains in the microstructure of a polycrystalline FE material with an enhanced electromechanical coupling from both a textured ceramic and a single crystal.

To arrive at an optimum texture of the ferroelectric polycrystal at which the material exhibits maximum piezoelectric performance as a hydrostatic transducer material, a global optimization method has to be employed (Jayachandran et al., 2010). Stochastic optimization techniques like simulated annealing (SA) are quite suitable in this respect as the objective function is not sensitive to the starting point of the iterative process. Besides being insensitive to the starting point, SA can search a large solution space and they can escape local optimum points thanks to the freedom for occasional uphill moves. Kirkpatrick et al. (1983) first proposed simulated annealing as a powerful stochastic optimization technique. The complex structure of configuration space is treated analogous to the state of material controllable by an adjustable parameter, the temperature, in simulated annealing. In other words, annealing is a strategy by which an optimum state can be approached by controlling the temperature. Annealing involves heating the material matrix to high temperature and then let it be cooled slowly so that at each step a near thermal equilibrium is achieved and finally render the material to a stable minimum energy crystalline (ordered) state. The idea to explore analogy of the annealing used in solid state physics with the optimization problems gives rise to simulated annealing technique. A control parameter similar to the temperature in physical annealing is introduced in optimization which will dictate the number of states to be accessed in going through the successive steps of the optimization algorithm before being settled in the minimum energy state (the optimum configuration).

2 MODEL SETUP

As remarked above, the crystallographic grains (crystallites) in an as-grown polycrystal are randomly oriented and require three angles to describe its orientation with reference to a fixed Cartesian coordinate system. Euler angles (φ, θ, ψ) can completely specify the orientation of the crystallographic coordinate system embedded in crystallites and thereby the orientation of crystallites (Goldstein, 1978).

2.1 Objective function and design variables

The parameters of the distribution viz., μ and σ the mean and standard deviation of the probability distribution function (pdf) characterise the Gaussian distribution,

$$f(\alpha|_{\mu,\sigma}) = \frac{1}{\sigma\sqrt{2\pi}} \exp - \frac{(\alpha-\mu)^2}{2\sigma^2} \tag{1}$$

Here α stands for the Euler angle (φ, θ, ψ). In the present work, parameters μ and σ perform the role of the control parameters which will decide the scatter of the orientations (Euler angles) and hence be critical to the piezoelectric response of the polycrystalline ferroelectric material. Hence μ and σ are the design variables of the optimization problem. Thus we are aiming to find an optimum set of these parameters from a solution space controlled by the laws of coordinate transformations from a crystallographic coordinate system embedded in the grains to a local coordinate system which coincides with the global frame of reference. Also, the solution space is bounded by distribution parameters μ and σ ranging from those of uniform (in the case of random polycrystal) to those of Gaussian distribution (in the case of poled polycrystal). A fairly uniform kind of distribution can be achieved by putting standard deviation (σ) equals 5 and for a poled ceramic ferroelectric the σ is set near zero.

The composite piezoelectric materials found useful application in transducers for hydroacoustic applications (Smith 1989; Gibiansky and Torquato, 1997]. There are several requirements for the piezoelectric used in these transducers. A major requirement for a sensitive transducer is that the piezoelectric material used in the composite must efficiently convert between electrical and mechanical energy. Effective piezoelectric constants and hydrostatic figures of merit such as the hydrostatic charge response dh, and hydrostatic coupling constant k_h manifest the piezoelectric sensitivity in hydroacoustic applications among other parameters (Gibiansky and Torquato, 1997; Topolov and Panich, 2009). In single crystalline materials like $BaTiO_3$ the piezoelectric strain shows a maximum when they are poled along a nonpolar axis Wada et al. 1999; Jayachandran et al., 2008). However, our objective is to search possible ways of enhancing the piezoelectricity in ceramic ferroelectric materials making it suitable for hydroacoustic applications. Given the difficulties in synthesizing good quality single crystals of fairly large size for integration and also the non-reliability of reproduction, polycrystals are often preferred to single crystals in device applications. In polycrystalline piezoelectric materials, the state of strain is inhomogeneous. Understanding the local and global ferroelectric response of these topologically complex materials by combining mathematical modeling and simulation could help effectively engineer material configurations and judicious selection of materials. The grain distribution parameters chosen by the simulated annealing (SA) algorithm will prompt a normal random generator thereby create a set of Euler angles (φ, θ, ψ). These Euler angles will dictate the coordinate transformation in the electromechanical property tensors appearing in the homogenization equations.

2.2 Homogenization of piezoelectric material

We have used mathematical homogenization method for the evaluation of the equilibrium, effective (macroscopic) piezoelectric and mechanical properties of a polycrystalline ferroelectric material possessing the lowest crystallographic symmetry. Further, the numerical solution of the coupled piezoelectric problems is sought using the finite element method (FEM) to eventually compute the homogenized coefficients. The mathematical theory of the homogenization method accommodates the interaction of different phases in characterizing both the macro- and the micromechanical behaviors (Sanchez-Palencia, 1980). In homogenization theory it is usually assumed that the material is locally formed by the spatial repetition of very small microstructures, when compared with the overall macroscopic dimensions. Further, it is assumed that the material properties are periodic functions of the microscopic variable, where the period is very small compared with the macroscopic variable. This assumption enables the computation of equivalent material properties by a limiting process wherein the microscopic cell size is approaching zero (Guedes and Kikuchi, 1990). The finite element method used for this study correlates each randomly oriented grain in a polycrystalline material with each element of the finite element mesh. Each grain in a polycrystalline material is assumed to be made of a single, pinned, chemically homogeneous ferroelectric domain.

The asymptotic analysis and homogenization of the piezoelectric medium [Telega, 1990; Nelli Silva et al., 1999) has resulted in the macroscopic piezoelectric coefficients

$$e^H_{prs}(\boldsymbol{x}) = \frac{1}{|Y|}\left\{\int_Y \left[e_{kij}(\boldsymbol{x},\boldsymbol{y})\left(\delta_{kp} + \frac{\partial R^{(p)}}{\partial y_k}\right)\left(\delta_{ir}\delta_{js} + \frac{\partial \chi_i^{(rs)}}{\partial y_j}\right) - e_{kij}(\boldsymbol{x},\boldsymbol{y})\frac{\partial \Phi_i^{(p)}}{\partial y_j}\frac{\partial \psi^{(rs)}}{\partial y_k}\right] dY\right\} \quad (2)$$

in index notation. Here e_{kij} are the piezoelectric stress coefficients of the single grain and a number of such distinct grains constitute the unit-cell. $\chi_i^{(rs)}$ is a characteristic displacement, $R^{(p)}$ is a characteristic electric potential, $\Phi_i^{(p)}$ and $\psi^{(rs)}$ are characteristic coupled functions and Y is the size of the unit-cell. δ is the Kronecker delta symbol. Also in contracted notation e_{kij}^H can be written as $e_{k\mu}^H$ where i, j, k =1,2,3 and μ = 1,2,..6 in the general three-dimensional case. The functions e_{kij}^H in Eq. (2) can be described in microscopic coordinate system y_i using the components of Euler transform tensors from crystallographic coordinates. The homogenized electromechanical properties which are the objectives of this study are the following: The piezoelectric strain coefficients,

$$d_{i\nu}^H = \sum_{\mu=1}^{6} e_{i\mu}^H s_{\mu\nu}^H \tag{3}$$

where $s_{\mu\nu}^H$ are the homogenized elastic compliance tensor. The hydrostatic charge response,

$$d_h^H = d_{33}^H + 2d_{31}^H \tag{4}$$

and the hydrostatic coupling coefficient

$$k_h^H = d_h^H / \sqrt{\in_{33}^T s_h^E} \tag{5}$$

where $\in_{33}^T$ is the unclamped (zero stress) dielectric permittivity and s_h^{EH} is the homogenized hydrostatic compliance defined by,

$$s_h^E = 2(s_{11}^E + s_{12}^E + 2s_{13}^E) + s_{33}^E \tag{6}$$

The piezoelectric response is determined along an arbitrary crystallographic direction determined by the Euler angles with respect to the reference frame of the unit-cell. The superscript *H* would be dropped henceforth from the homogenized coefficients for brevity.

The numerical model for this work is developed in 3D incorporating the anisotropy of the polycrystalline ferroelectric at the grain level. The microscopic system of equations is computationally solved using the FEM. Since our model is able to predict the macroscopic electromechanical coefficients of all piezoelectric crystal classes, application of the same to ferroelectric $BaTiO_3$ requires the knowledge of point group symmetry which is tetragonal 4*mm*. The unit-cell is discretized using a mesh of 15×15×15 finite elements and each node of the trilinear solid element is allowed four degrees of freedom (one electric potential and three displacements). Full integration (2-point Gaussian integration rule in each direction) is used for the evaluation of the stiffness, piezoelectric and dielectric matrices and for the homogenization. As the representative unit-cell is expected to capture the response of the entire piezoelectric system, particular care is taken to ensure that the deformation across the boundaries of the cell is compatible with the deformation of adjacent cells. Hence all the load cases are solved by enforcing periodic boundary conditions in the unit-cell for the displacements and electrical potentials. The numerical simulation of ceramic $BaTiO_3$ are done using the parameters of single crystal data from Zgonic et al. (1994) using the present homogenization model computationally implemented in *Fortran*.

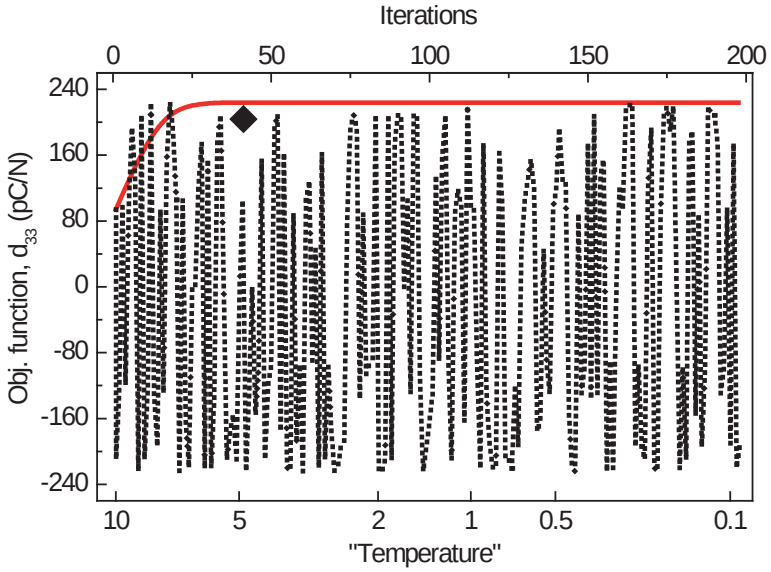

Figure 1: Energy" (piezoelectric coefficient d_{33}) as a function of *temperature* in ferroelectric single crystal $BaTiO_3$. The diamond indicates the experimental value obtained by Wada et al. (1999) for [111] oriented single crystals

2.3 Stochastic Optimization

The basic concept of simulated annealing (SA) is based on the Metropolis algorithm (Metropolis et al., 1983) for simulating the behavior of an ensemble of atoms that are cooled slowly from their melted state to the minimum energy ground state. In order to apply SA to a piezoelectric material, we must first introduce the notion of ''system energy.'' In order to be consistent with our definition of design variables, let.

$$E(R_i) \equiv \tau_{i\mu}(\alpha) \equiv \tau_{i\mu}(\sigma, \mu) \tag{7}$$

be the surrogate for energy of a particular configuration R_i. Here $\sigma \in [0,5]$ and $\mu \in [0, \frac{\pi}{2}]$. The distribution parameters are selected from a randomized set of μ and σ.

The ultimate goal of SA is to find the ground state(s), i.e., the minimum energy configuration(s), with a relatively small amount of computation. The likelihood that a configuration, R_i, is allowed to exist is equal to the Boltzmann probability factor $P(R_i) = \exp\left[-E(R_i)/k_B T\right]$ where k_B is the Boltzmann constant and T is the temperature.

The optimization problem can be summarized as to find (γ) that maximize

$$\left.\begin{array}{l} f(\gamma) \equiv \tau_{jv}(\gamma) \\ \text{subject to} \\ : -\pi \le \gamma \equiv (\phi, \theta, \psi) \le \pi \big\} \text{ for single crystals} \\ : 0 \le \gamma \equiv (\mu_\phi, \mu_\theta, \mu_\psi) \le \pi/2 \\ : 0 \le \gamma \equiv (\sigma_\phi, \sigma_\theta, \sigma_\psi) \le 5 \end{array} \right\} \text{for polycrystals} \tag{8}$$

The optimization (implementation in *Matlab* runs on a standard desktop PC) algorithm calls homogenized objective functions at every run.

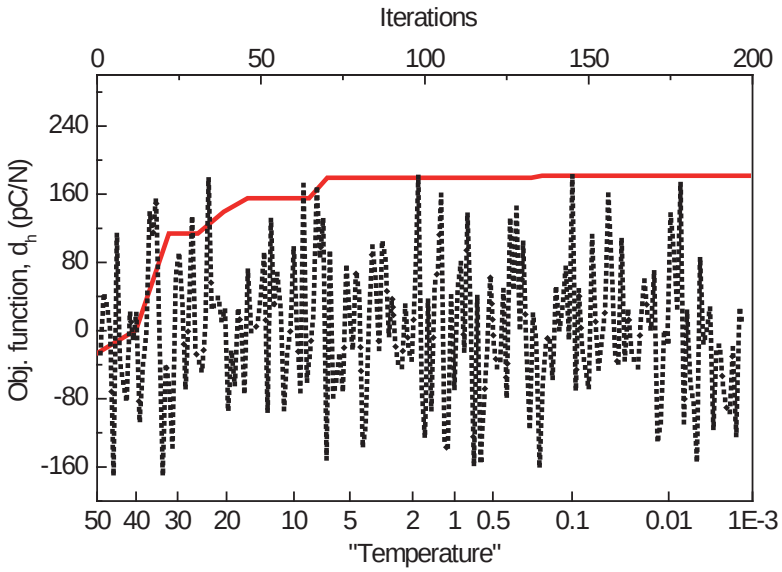

Figure 2: "Energy" (hydrostatic charge response d_h) as a function of *temperature* and number of iterations in ferroelectric single crystal $BaTiO_3$

3 RESULTS AND DISCUSSION

As stated above we have six design variables, *viz.* $\sigma_\varphi, \mu_\varphi, \sigma_\theta, \mu_\theta, \sigma_\psi$ and μ_ψ that corresponds the standard deviations and means of the Euler angles (φ, θ, ψ) expressed in radians. The *temperature* T is set to fall by 20% from each of the previous step, i.e., $T_{k+1} = 0.8T_k$.

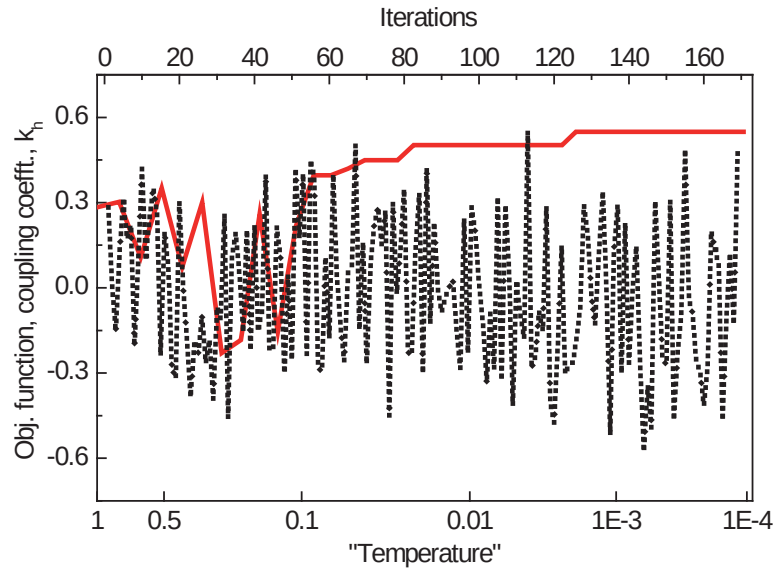

Figure 3: Energy" (hydrostatic coupling coefficient k_h) as a function of *temperature* and number of iterations in ferroelectric single crystal $BaTiO_3$

First we study the piezoelectricity $BaTiO_3$ single crystal. The evolution of the objective function d_{33} with the temperature is shown in Fig.1. The piezoelectric coefficient d_{33} obtained after optimization is 223.7 pC/N which compares exactly with our homogenization results reported recently (Jayachandran et al., 2009). The solution (φ, θ, ψ) is (-2.182, 0.873, -0.175). This corresponds to $(\phi = -125^\circ, \theta = 50^\circ, \psi = -10^\circ)$. This is one of the parallel <111> directions of the $BaTiO_3$ single crystal along which the maximum piezoelectric coefficient of d_{33} = 203 pC/N is measured by Wada et al. (1999).

Fig. 2 illustrates the results of optimization corresponding to the hydrostatic charge response d_h in $BaTiO_3$ single crystal. As shown in Eq. 4, this property depends on the transverse piezoelectric coefficient (which in general is negative) d_{31}. Hence it would be certainly less than the maximum d_{33} one can get from the system. Here it attains a maximum of d_h.= 181.8 pC/N at the optimal solution of $(\phi = 110^\circ, \theta = 55^\circ, \psi = 0^\circ)$. This direction will also coincides with one of the <111> orientations of the $BaTiO_3$ single crystal. Next we study the optimization of electromechanical coupling k_h in single crystal $BaTiO_3$. Fig. 3 shows the convergence of the coupling coefficient k_h of single crystal $BaTiO_3$ with the number of iterations and decreasing *temperature*. The optimum value is k_h = 0.55 at $(\phi = 75^\circ, \theta = 40^\circ, \psi = 5^\circ)$.

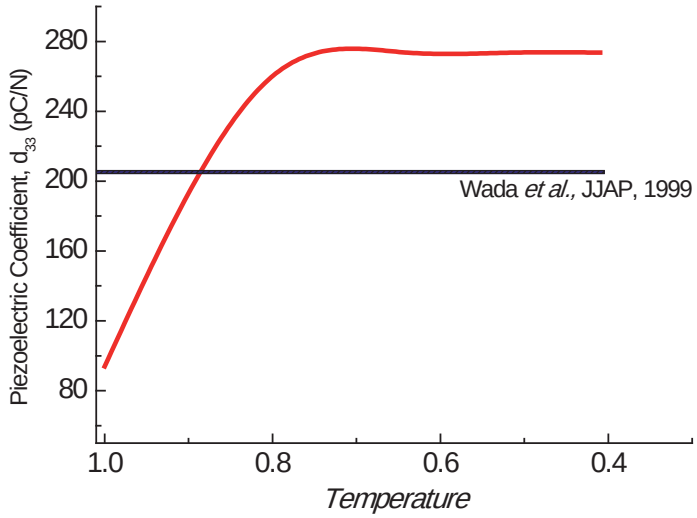

Figure 4: Energy" (Piezoelectric coefficient d_{33}) as a function of *temperature* in ferroelectric polycrystal $BaTiO_3$. The horizontal line gives the experimental value for [111] oriented single crystal from Wada et al. (1999).

Next, we apply this optimization procedure to the ferroelectric polycrystals. We analyze the case with $(\sigma_\phi, \sigma_\theta, \sigma_\psi) \in [0,\ 5]$ and $(\mu_\phi, \mu_\theta, \mu_\psi) \in [0, \frac{\pi}{2}]$. The results are shown in Fig. 4. The solution $(\sigma_\phi, \mu_\phi, \sigma_\theta, \mu_\theta, \sigma_\psi, \mu_\psi)$ obtained is (4.7, 0.873, 0, 0.698, 1.8, 1.223). The objective function converges with a value d_{33} = 273.7 pC/N which is very much higher than both [001] poled and [111] poled single crystals. This supplements the notion that randomness in the orientation of grains, if utilized judiciously, might be useful for manufacturing ceramics with better piezoelectric performance than single crystals. The solution suggests to keep the Euler angle related to the orientation of ab-plane of the crystallites (φ and ψ) to be kept in random

rather than keeping their value at zero while the orientation θ, of c-axes is kept at 0.698 radians (θ = 40°). Such a configuration of the crystallites in a ceramic FE would result in a better piezoelectric than an oriented single crystal.

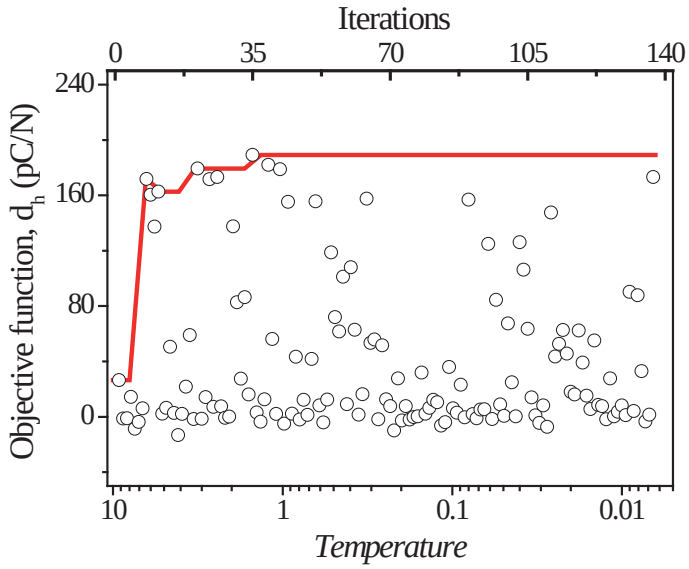

Figure 5 Hydrostatic charge response d_h as a function of *temperature* and number of iterations in ferroelectric polycrystal $BaTiO_3$

Fig. 5 depicts the optimization results of hydrostatic charge response d_h of the polycrystal ferroelectric $BaTiO_3$. Here the optimum value obtained is d_h = 189.20 pC/N with a solution $(\sigma_\phi, \mu_\phi, \sigma_\theta, \mu_\theta, \sigma_\psi, \mu_\psi)$ equal to (0, 0, 0.6, 0.087, 0). This value of d_h is higher than the single crystal maximum charge response of d_h.= 181.8 pC/N. In summary, tailoring the microstructure by tuning the texture of the ferroelectrics would result in materials possessing piezoelectric properties better than single crystals. In view of the difficulties associated with the manufacture of the single crystals, the present result would bear importance in applications involving piezoelectricity.

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