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Full Length Research Paper

Artificial-Neural-Network modeling of the compressive uniaxial stress dependence of ferroelectric hysteresis: An application to soft lead zirconate titanate ceramics

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In this work, artificial neural network (ANN) modeling was used to model ferroelectric hysteresis under the influence of compressive uniaxial stress using the hysteresis data obtained from soft lead zirconate titanate as an application. The main objective is to model the role of external stress, including electric field perturbation, on the complex hysteresis properties, which are hysteresis area, remnant polarization, coercivity and loop squareness. With its false tolerance abilities, ANN was used to predict how the stress direction (on applying and releasing), the stress magnitude (σ) the electric field amplitude (E_0), and the electric frequency (f) affect on the hysteresis properties, quantitatively. The best network architecture with highest accuracy was found in the ANN training through extensive architecture search. It was then used to predict hysteresis properties of the unseen testing patterns of input. The predicted and the actual testing data were found to match very well for the whole extensive range of considered input parameters. This well match, even when the stress was applied, certifies the ANN one of the superior techniques, which can be used for the benefit of technological development of ferroelectric applications.

Key words: Artificial neural network, hysteresis properties, soft lead zirconate titanate, uniaxial stress.

INTRODUCTION

During recent years, the ferroelectric hysteresis topic has become of frequent investigating issue due to the need of important ferroelectric applications (Auciello et al., 1998; Uchino, 2000). In such an application, the amplitude (E_0) and frequency (f) dependence of hysteresis parameters are of important consideration. Both experimental and theoretical studies have mainly focused on the use of power law scaling to investigate how hysteresis properties response to external field parameters in a form of $A \propto f^{\alpha} E_0^{\beta}$ where A denotes the hysteresis area, α and β are exponents to the scaling, for example, consider Uchino (2000). In spite of its reasonable success on finding how hysteresis area relates to the field, each exponent obtained in this way is not truly independent. Therefore, in previous works, α and β were extracted separately (Yimnirun et al., 2006a). Specifically, one exponent was extracted at a time and when it was retrieved, it was assumed constant and fed back to the power law to find another exponent. However, though this method is sound, the extracted exponents are very

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Abbreviations: **ANN**, Artificial neural network; **PZT**, lead zirconate titanate; σ, stress magnitude; E_0 the electric field amplitude; f, frequency; **MLP,** multilayer perceptron; **BP,** back propagation; A, loop area; P_r, remnant polarization; E_c, hysteresis coercivity , S, hysteresis loop squareness; **MAE,** mean absolute error.

vulnerable where a small change or error in α could cause a considerable change in β. In addition, with further including of relevant parameters in the scaling such as the inclusion of the stress parameter σ to seek for the scaling in the form $A \propto f^{\alpha} E_0^{\beta} \sigma^{\gamma}$, the problem becomes even more complicated and some approximation has to be applied. For instance, instead of the sole magnitude of the hysteresis area, one has to consider the difference between the area at current applied stress and that of the unstressed to form the scaling (Yimnirun et al., 2006b).

$$\left\langle A - A_{\sigma=0} \right\rangle \propto f^{\alpha} E_0^{\beta} \sigma^{\gamma} . \quad (1)$$

Though its reasonable success in constructing the scaling formalism, it is very obvious that the zero-stress hysteresis-area $A_{\sigma=0}$ in Equation (1) must be known before hand for each f and E_0 conditions. On the other hand, in some systems, the unstressed condition is not accessible such as in films structure where the internal stress is induced from the lattice spacing mismatch between the films and the substrate. In this case, one cannot make the best use of Equation (1) in the modeling. Instead, one has to phenomenologically propose new scaling function using either trial-and-error or more sophisticate empirical methods. Moreover, when including the minor hysteresis loop, 2 scaling functions have to differently proposed for minor loop and saturated loop even without the stress and in the same ferroelectric ceramic (Wongdamnern et al., 2009). Further, the scaling exponents are not truly constants but a function of field parameters, for example, α may be a function of E_0 and β may be a function of f (Wongdamnern et al., 2009). Therefore, there is no guarantee if there really exists simple power-law-scaling form (where the exponents α, β and γ are truly constant) for all ferroelectric systems. In such cases, the simple power-law-scaling is no longer simple.

Consequently, in this work, artificial neural network (ANN) which is another sophisticate technique was applied to model the hysteresis behavior. ANN is a technique widely used in industries for various purposes due to its ability to 'learn' from examples. For instance, ANN was used in modeling concrete strength (Hakim et al., 2011), landslide risk analysis (Pradhan and Lee, 2009), traffic accidents (Bayata et al., 2011), faults in software systems (Ardil and Sandhu, 2010), rainfall-runoff prediction (El-Shafie et al., 2011), etc. Further, the ANN was recently found to be useful in modeling properties of material prepared/measured under various conditions (Laosiritaworn, 2008; Laosiritaworn et al., 2008, 2010a, 2010b, 2011; Lemine et al., 2010; Laosiritaworn and Laosiritaworn, 2009; Laosiritaworn and Chotchaithanakorn, 2009). Therefore, in this work, the ANN was used to model ferroelectric hysteresis under mechanical loading condition using soft Lead Zirconate Titanate as an application.

BACKGROUND THEORIES

ANN is a statistical model of actual system built by tuning a set of parameters known as weight. It can perform function mapping for a set of given value of inputs to corresponding set of outputs (Dayhoff, 1990; Swingler, 1996). ANN simulates biological neural networks in human brains so that it can learn to pick up relationship or pattern in data the same way human brain function. The type of ANN used in this paper is a multilayer perceptron (MLP) which consist of input layer, hidden layer and output layer (Figure 1). Each layer consists of simple processing elements called neuron and neurons in each layer are connected together to form a neural network. Weight is assigned to each connection between neurons, initially by small random number. By tuning adjusting these weights, ANN can be used to learn relationship between input and output.

A number of training algorithms are available for weight tuning process. In this study, the back propagation (BP) algorithm, one of the most widely used algorithms (Swingler, 1996), was applied. In BP learning, two steps were performed, the forward pass and the backward pass. In the forward pass, inputs are fed to ANN. Each neurons attain output by calculate weighted sum (S_j) from $\sum_i a_i w_{ij}$, where a_i is the activation level of unit i, and w_{ij} is the weight from unit i to unit j. Then, the logistic transfer function, that is, $g(x) = \frac{1}{1+e^{-x}}$ where $x = S_j$, were applied to the output. Then, $g(x = S_j)$ becomes the output of unit j, and the same procedure repeats for all neurons to obtain the final output. This output is then compared with its corresponding target value and the deviation between them is calculated in the backward pass. Error in the output layer is calculated from $\delta_j = (t_j - a_j) g'(S_j)$ and $\delta_j = \left[\sum_k \delta_k w_{kj} \right] g'(S_j)$ for the hidden layers. In these equations, t_j is the target value for unit j, a_j is the output value for unit j, $g'(x)$ is the derivative of the logistic function g and S_j is weighted sum of inputs to j. Then, the weight adjustment is calculated as $\Delta w_{ji} = \eta \delta_j a_i$ where η is the learning rate. These forward and backward processes repeat with new input vector until stopping criteria are met (Nascimento et al., 2000).

METHODOLOGY

In this work, the ferroelectric hysteresis data of the soft lead

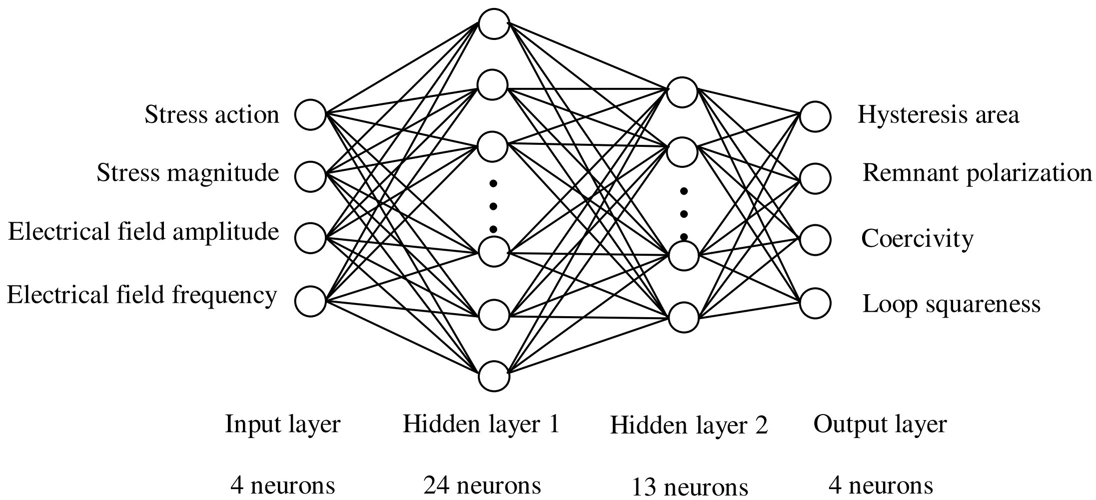

Figure 1. The schematic diagram of the ANN used in this work.

Table 1. Input and output data used in ANN training.

Parameter	Type	Data format	Max	Min
Stress action	Input	Categorical	n/a	n/a
Stress magnitude	Input	Numerical	0.75	0
Field amplitude	Input	Numerical	1800	600
Field frequency	Input	Numerical	100	0.6
Hysteresis area	Output	Numerical	49111000	248720
Remnant polarization	Output	Numerical	8496.2	147.3
Hysteresis Coercivity	Output	Numerical	1654.9	169.34
Loop squareness	Output	Numerical	0.53345	0.10709

zirconate titanate (PZT) ceramic was used to train the network. The hysteresis loops were measured at room temperature (25°C) from commercial soft PZT ceramic disks (PKI-552, Piezo Kinetics Inc., USA) with diameter of 10 mm and thickness of 1 mm with f covering from 2 to 100 Hz and E_0 from 600 to 1800 mV. Details of the measuring system were described elsewhere (Yimnirun et al., 2006b, 2006c). Then, from the hysteresis loops, the loop area A, the remnant polarization P_r, the hysteresis coercivity E_c, and the hysteresis loop squareness S were extracted and used as output hysteresis data for the ANN training. The input data for the ANN are the field frequency f, the field amplitude E_0, the stress application (applying or releasing), and the stress magnitude σ. These actual input experimental data were used to train the artificial neural network for predicting the output hysteresis properties. Input and output of the ANN are summarized in Table 1.

RESULTS AND DISCUSSION

As can be seen in Tables 1 and 2 network architectures were considered to maximize the training efficiency and for accuracy comparison. Specifically, in the first architecture, a single ANN was used to model four outputs in the same time. Therefore, there were 4 neurons in input layer (representing stress application, σ, E_0, and f), 4 neurons in output layer (which are A, P_r, E_c and S representing hysteresis area, remnant polarization, hysteresis coercivity, and loop squareness respectively). In the second architecture, 4 ANNs were trained separately to model the four outputs, that is, 1 network for 1 output). Then the number of hidden layers and hidden nodes in each hidden layers search were conducted for up to 2 layers and up to 30 neurons in each layer. Appropriate number of hidden layer and hidden nodes were achieved through heuristic and exhaustive search. Note that the best architecture is listed in the format XX-XX-XX-XX where XX refers to the number of nodes in input layer, first hidden layer, second hidden layer and output layer respectively. After that, the raw input-output data of 788 records were separated into 3 sets which are training, validate and testing dataset at the ratio of 536: 126: 126, respectively.

From the training, the networks with highest accuracy for each architecture were found and they are summarized in Table 2. The network accuracy is measured in terms of mean absolute error (MAE) and the square of

Table 2. ANN training results. The symbols A, P_r, E_c and S refer to hysteresis area, remnant polarization, coercivity and loop-squareness respectively, whereas MAE and r^2 refer the mean absolute error and square of correlation coefficient.

ANN architecture		ANN output	Training		Testing	
			MAE	r^2	MAE	r^2
First	4-24-13-4	A	748556.95	0.971936	920018.07	0.963404
		P_r	213.03	0.940677	250.451121	0.906773
		E_c	34.263541	0.975414	35.135949	0.980771
		S	0.009821	0.910215	0.012338	0.879406
Second	4-19-25-1	A	561048.86	0.986167	767065.37	0.978375
	4-22-22-1	P_r	157.87	0.968957	227.54	0.933713
	4-30-27-1	E_c	24.66	0.988093	33.61	0.983918
	4-30-20-1	S	0.006037	0.969711	0.011621	0.897849

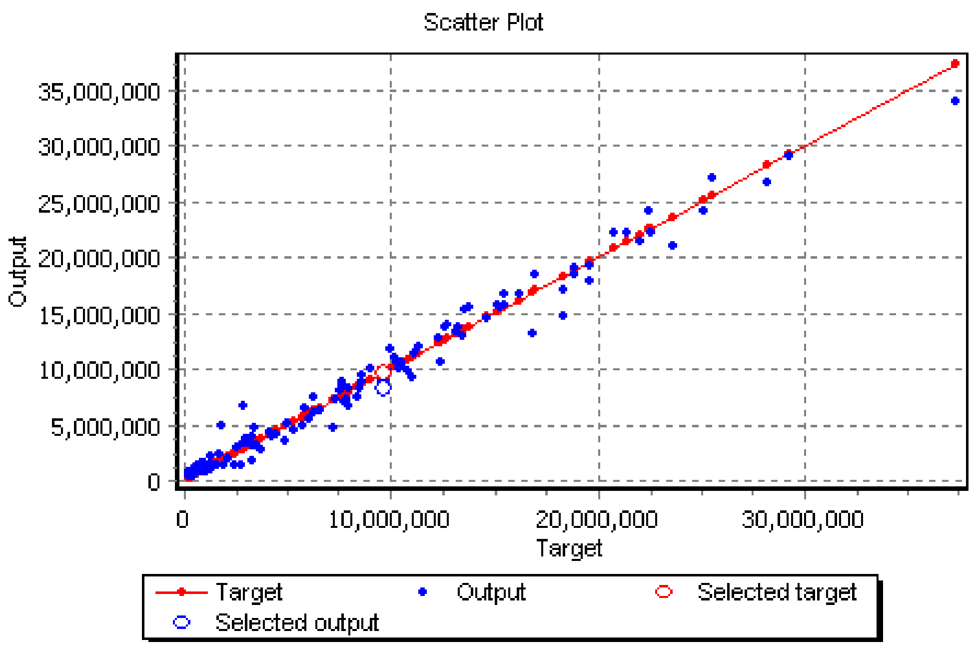

Figure 2. Scatter plot of target (testing group) and output of hysteresis area output generated from the area architecture (4-19-25-1).

square of correlation coefficient (r^2), that is,

$$MAE = \frac{\sum_{i=1}^{n} |f_i - y_i|}{n} \quad \text{and}$$

$$r = \frac{n\sum_{i=1}^{n} f_i y_i - \left(\sum_{i=1}^{n} f_i\right)\left(\sum_{i=1}^{n} y_i\right)}{\sqrt{n\left(\sum_{i=1}^{n} f_i^2\right) - \left(\sum_{i=1}^{n} f_i\right)^2}\sqrt{n\left(\sum_{i=1}^{n} y_i^2\right) - \left(\sum_{i=1}^{n} y_i\right)^2}}$$

where f_i is the prediction from neural network for record i, y_i is the actual value for record i, and n is the total number of data. In general, the smaller of MAE and the closer of r^2 to 1 are desirable. Therefore, from Table 2, it can be concluded that training with 4 separate networks (second architecture) can improve modeling accuracy judging from both MAE and r^2. However, the first architecture required much less time and effort in training and provides acceptable r^2 (> 0.8978). An example of scatter plot can be found in Figure 2, which shows the plot between target value (testing group) and output from ANN of the network trained to model hysteresis area with the architecture of 4-19-25-1. Further, Figure 3 shows the comparison between the actual data (open square) and that from the ANN predicting (lines). Being evident, the

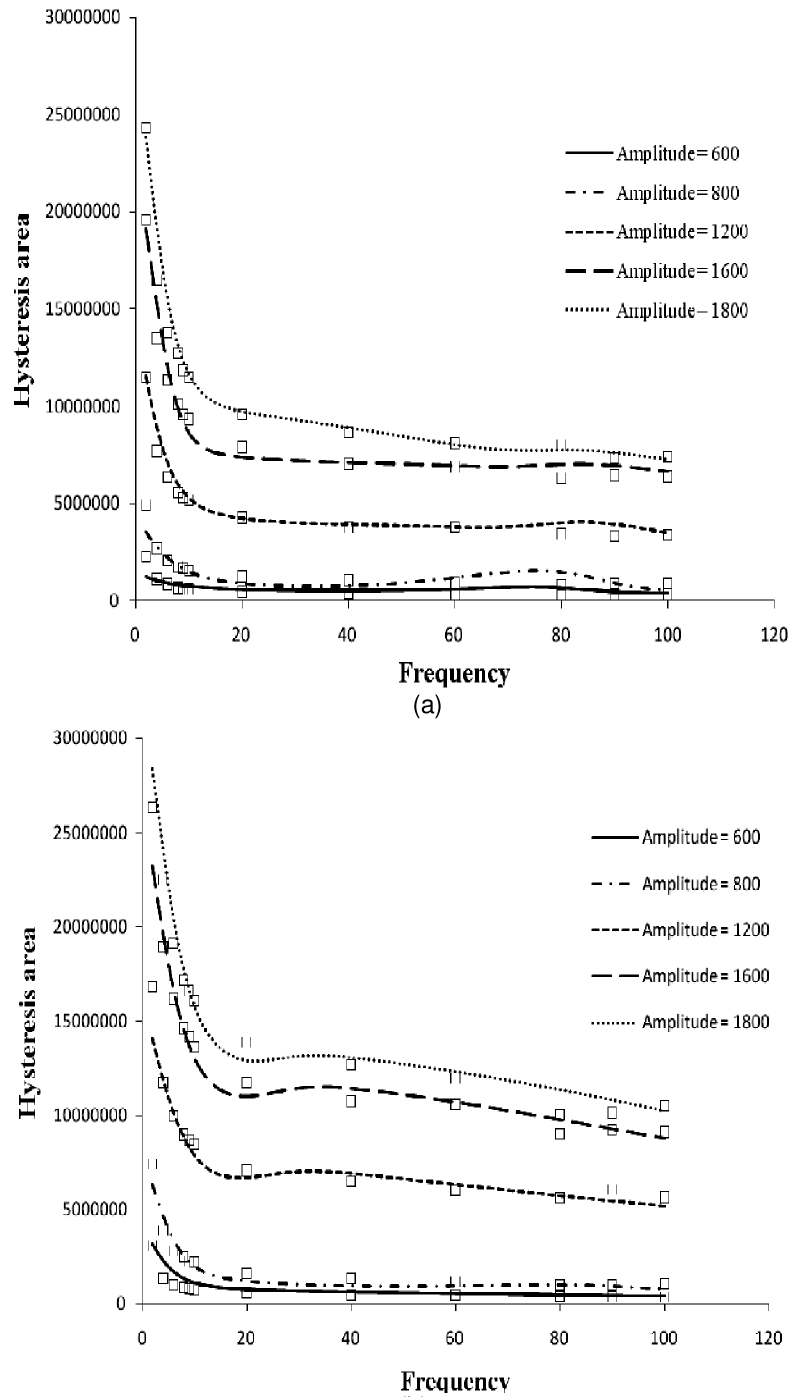

Figure 3. Comparison of the actual data (open square) and that from the ANN predicting (lines) for the field amplitude E_0 ranging from 600 to 1800 mV and at σ = 0.375 kN/π(5mm $)^2$ for (a) stress applying and (b) stress releasing.

predicting data forms well representatives for the actual experiments data for both applying and releasing stresses. In addition, unlike the previous power-law-scaling investigation where the scaling was performed on one particular stress application (applying) (Yimnirun et al., 2006b), as applying and releasing stress are of different behaviors, this work can modeling both stress applying and releasing at the same time. Therefore, including with the good r^2 provided, it can be concluded that the ANN is one of the appropriate and successful techniques in modeling ferroelectric hysteresis even under both electrical and mechanical perturbations.

It is also of interest to compare, discuss, pin-point, and summarize the benefit of using ANN upon previously used techniques in the analysis of the hysteresis data. Traditionally, the most commonly used technique in the investigating of how hysteresis properties depend on the external perturbation (that is, field parameters, temperature, external stress, etc.) is the power law scaling technique. The reason behind using this empirical technique is that it is simple and easy to implement which could suit the purpose without the need to know fundamental knowledge of the considered system. Nevertheless, there do exist a main drawback as the power law scaling technique is very much depend on the complexity in the ferroelectric material (for example, films or bulk, hard or soft ferroelectrics, grain size distribution, porosity, etc.) and ranges of the external perturbation (for example, how high is the measuring temperature (above or below critical temperature), how large is the field amplitude (sub-coercive field or saturated field), how fast is the field switching, how high is the stress acting on the materials and what is the applied stress direction (compressive or tensile), etc.). Therefore, with these numerous degrees of freedom, the simple power law scaling technique is no longer simple. Researchers had to perform the scaling with many approximations and had to propose limitation on the allowed ranges of the applied perturbation. This is unless it may be not possible to perform or obtain appropriate fit on the measured hysteresis data. For instance, even with the same piece of ferroelectric ceramics, increasing and decreasing stresses yield different scaling functions and hence difference the exponents to the power law function (Yimnirun et al., 2006b). Therefore, this is evident that the traditional power law technique is not applicable when the problems become very complex such as under the supplied dynamic field and with mechanical loading. Further, as previous investigation could perform scaling only for the difference in hysteresis area (between those at finite stress and zero stress), not the absolute area (Yimnirun et al., 2006b), the knowledge in predicting hysteresis behavior is somewhat limited.

On the other hand, the ANN technique proposed and used in this work tends to overcome the difficulties encountered by the traditional power law technique. This is due to the ability in ‘learning by experiences’ of the ANN which could correctly predict the hysteresis behavior even under the loading condition during either applying or releasing stresses. Another benefit of using this ANN upon the scaling technique is that the ANN has to some extent high tolerance to electrical noises, generally arisen from poor experiment setup. These noises result in some data points with very high error, which have to be removed before performing the scaling unless the fit might not converge. Nevertheless, if the error is symmetry due to the randomness of the noise or the noise affects only minor parts of the data signal, the ANN is not strongly affected by these noises and can still be used in the modeling. As a result, for example, in this work, all of these illustrate on how the use of the ANN in hysteresis modeling leads to significant improvement over traditional power law modeling.

Further, to enhance the use of this ANN technique on modeling ferroelectric hysteresis, the next stage will be carried on modeling hysteresis properties from materials with possible different crystal structures depending on the chemical composition. For instance, in some mixed ferroelectrics, which are prepared from two or more different ferroelectric materials, could have different crystal structures depending on the ratio of those different ferroelectric compositions, and this leads to different ferroelectric properties (Uchino, 2000). Therefore, in the future work (research direction), the ANN will be used in modeling dynamic hysteresis properties to predict how they depend on microscopic crystal structures, chemical composition and external electrical field parameters in a particular material.

CONCLUSION

In this work, the ANN was used to model the hysteresis properties of soft PZT ceramics under loading condition. Based on the agreement between the actual experiment values and those from the ANN prediction, the ANN has assured itself one of the fruitful techniques in modeling ferroelectric hysteresis properties even under applied stresses. Further, without the need to separate the data for the stress applying and stress releasing, the ANN investigation further approved its advantage over the conventional power law scaling technique.

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