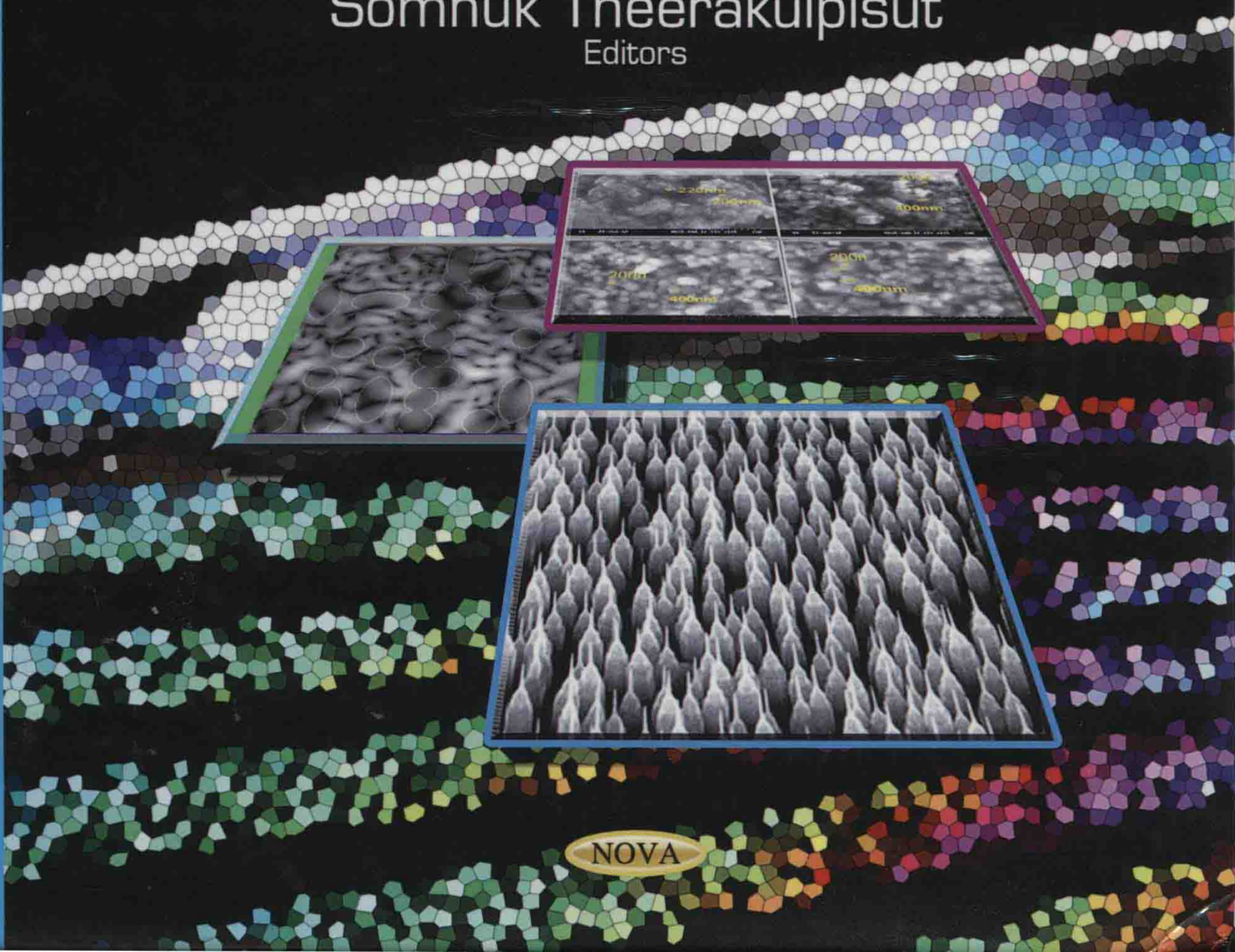
Nanotechnology Science and Technology

Advanced Materials

Studies and Applications

Ivan A. Parinov • Shun-Hsyung Chang
Somnuk Theerakulpisut
Editors



NANOTECHNOLOGY SCIENCE AND TECHNOLOGY

ADVANCED MATERIALS

STUDIES AND APPLICATIONS

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SHUN-HSYUNG CHANG
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NANOTECHNOLOGY SCIENCE AND TECHNOLOGY

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PREFACE

The advanced materials and their applications based on nanotechnology and piezoelectric approaches have found a tremendous interest in the modern science and techniques. The book presents processing techniques, physics, mechanics, and applications of novel materials. The book concentrates on some nanostructures, ferro- and magnetoelectric crystals, materials and composites, materials for solar cells, polymeric composites etc. There are present nanotechnology approaches, modern piezoelectric techniques, and also studies of the structure-sensitive properties of the materials. Great attention is devoted to novel devices with high accuracy, longevity and extended possibilities to work into wide temperature and pressure ranges, etc., which show characteristics defined by used materials and composites with improved properties opening new possibilities in study of various physical processes, in particular transmission and receipt of signals under water.

This collection of 30 papers presents selected reports of the 2014 International Symposium on "Physics and Mechanics of New Materials and Underwater Applications" (PHENMA-2014), which has been taken place in Khon-Kaen, Thailand, 27-29 March, 2014 (http://phenma2014.math.sfedu.ru), devoted to the 50th anniversary of Faculty of Engineering, Khon Kaen University and sponsored by the Russian Foundation for Basic Research, National Science Council of Taiwan, South Scientific Center of Russian Academy of Sciences, New Century Education Foundation (Taiwan), Ocean & Underwater Technology Association, Unity Opto Technology Co., EPOCH Energy Technology Corp., Fair Well Fishery Co., Formosa Plastics Co., Woen Jinn Harbor Engineering Co., Lorom Group, Longwell Co., Taiwan International Ports Co., Ltd. and South Russian Regional Centre for Preparation and Implementation of International Projects.

The thematic of the PHENMA-2014 continues ideas of previous symposia PMNM-2012 (http://pmnm.math.rsu.ru) and PHENMA-2013 (http://phenma.math.sfedu.ru), whose results have been published in the edited books "Physics and Mechanics of New Materials and Their Applications", Ivan A. Parinov, Shun Hsyung-Chang (Eds.), Nova Science Publishers, New York, 2013, 444 p. ISBN: 978-1-62618-535-7 and "Advanced Materials - Physics, Mechanics and Applications". Springer Proceedings in Physics. V. 152. Shun-Hsyung Chang, Ivan Parinov, Vitaly Topolov (Eds.), Springer Heidelberg, New York, Dordrecht, London, 2014, 380 p. ISBN: 978-3319037486, respectively.

The presented papers are divided into four scientific directions: (i) processing techniques of advanced materials, (ii) physics of advanced materials, (iii) mechanics of advanced materials, and (iv) applications of advanced materials.

Into framework of the first theme are considered, in particular the search and development of smart materials, investigation of $NaTaO_3$ photocatalyst materials, studies of the doped ZnO thin films grown by dual-plasma-enhanced metal-organic chemical vapor deposition and pulsed laser deposition, characterization of ZnO nanocrystals, researches of sapphire laser processing and optimization of cure processing of polymeric composite.

The second direction covers structure and properties of spinel-perovskite lead-free magnetoelectric composites, finite-element modeling piezoelectric and magnetoelectric composites, porous piezoceramics and ceramic matrix composites, piezoelectric elements with inhomogeneous polarization, acoustic emission diagnostics of the kinetics of physicochemical processes in liquid and solid media, MDSM structures and thermoelectric capacitor with the semiconductor plate.

From viewpoint of mechanics in the third section are studied the problem on flat punch sliding on boundary of elastic half-plane, the coupled boundary-value problems of anisotropic elasticity, voscoelasticity, elastodynamics, poroelastodynamics and numerical methods (BIE and BEM) used for their study. Moreover, there are discussed the semi-inverted method in the problem of plastic deformation of cylindrical shell, and stress-strain state of layered anisotropic elliptic construction under action of impulse load.

The fourth direction demonstrates novel results in modeling and experimental studies of ultrasound transducers for medical applications, energy efficiency of piezoelectric generators, optic devices for measurement of displacements, based on method of control object highlighting by using laser interferometer, LEDs for marine applications, transmission signals under water, protection of equipment from excessive pressure and study of rigidity of the Novikov gearing teeth.

The book is addressed to students, post-graduate students, scientists and engineers taking part in R&D of nano-materials, ferro-piezoelectrics and other advanced materials and composites, presented in the book, and also participating in manufacture of different devices based on them having broad applications in different areas of modern science and technique, in particular in underwater communication.

October, 2014

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PART I. PROCESSING TECHNIQUES OF ADVANCED MATERIALS

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Chapter 1

SEARCH AND DEVELOPMENT OF SMART MATERIALS BASED ON SOLID SOLUTIONS OF SYSTEMS WITH DIFFERENT CHEMICAL COMPOUNDS

L. A. Reznichenko*[*]*, L. A. Verbenko, O. N. Razumovskaya, L. A. Shilkina, S. I. Dudkina, A. G. Abubakarov, A. V. Pavlenko, I. M. Aliev, S. Kh. Alikhadzhiyev, G. N. Tolmachev and H. A. Sadykov

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ABSTRACT

The structural and physical properties of $Pb(Ti, Zr)O_3 - \sum^4_{n=1}(PbB'_{1-\alpha}B''_\alpha O_3)_n$ solid solutions markedly different in electric hardness were correlated with the average electronegativity of the metal atoms on the *B* site (EN_B). The electric hardness of the solid solutions was shown to be proportional to EN_B (*B*-O bond covalence). The observed correlations can be used to predict properties of new piezoelectric ceramics.

Dielectric permittivity measurements in $Pb(Ti, Zr)O_3 - \Sigma^4_{n=1}(PbB'_{1-\alpha}B''_\alpha O_3)_n$ solid-solution systems demonstrate that the permittivity $\varepsilon^T_{33}/\varepsilon_0$ of the solid solutions correlates with the molar ratio from the more soft-electric to the less soft-electric component of $PbB'_{1-\alpha}B''_\alpha O_3(\gamma)$. With increasing γ, $\varepsilon^T_{33}/\varepsilon_0$ rises because of the decrease in the uniform cell distortion parameter. The permittivity is also sensitive to structural disordering at $\gamma >$ 1 and reduction in the density of the materials at $\gamma >> 1$.

The structural and dielectric ($\varepsilon^T_{33}/\varepsilon_0$) properties of $Pb(Ti, ZrO_3) - PbB'_{1-\alpha}B''_\alpha O_3 - PbB'''_{1-\beta}B''''_\beta O_3$ solid solutions were studied along the morphotropic region. Depending on the nature of the substituent cations, the properties of the solid solutions either vary monotonically with the content of the soft-electric component or exhibit extrema at a certain average electronegativity of the B-site cations.

Conditions for fabricating $(Ba_{0.5}Sr_{0.5})Nb_2O_6$ ceramics are optimized. It is shown that materials synthesized at $T = 1250$ °C and sintered at $T = 1375 - 1400$ °C exhibit the best ceramic characteristics. The $(Ba_{0.5}Sr_{0.5})Nb_2O_6$ solid solution is found to be a ferroelectric

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with a diffuse phase transition. The possibility of fabricating $(Ba_{0.5}Sr_{0.5})Nb_2O_6$ thin films from ceramic targets prepared in the resulting optimum regimes is shown.

Nonstoichiometric bismuth ferrite Bi_xFeO_3 ($0.88 \leq x \leq 1.04$) fabricated via solid-state synthesis is investigated by means of X-ray diffraction and electrical methods. The periodicity of the formation of impurity phases and variations in the structural parameters and densities of ceramic samples are established. The correlation between the number of impurity phases and characteristics of the dielectric spectra in the cation/anion-deficient ($0.88 \leq x < 1.00$) and cation/anion-excess ($1.00 < x \leq 1.04$) nonstoichiometry regions of Bi_xFeO_3 is confirmed. The observed effects are explained for the actual (defect) structure of bismuth ferrite.

1. Effects of B-Cation Electronegativity and B-O Bond Covalence on the Properties of Multicomponent Solid Solutions Based on Lead Zirconate Titanate

Earlier [1,2], we investigated the effect of the electronegativity of *B*-cations, EN_B, on the structural and electric properties of multicomponent solid solutions based on lead zirconate titanate (PZT),

$$Pb(Ti, Zr)O_3 - \sum^{4}_{n=1}(PbB'_{1-\alpha}B''_{\alpha}O_3)_n, \qquad (1)$$

containing roughly equal amounts of non-PZT components and close in Ti/Zr ratio. The results of those studies demonstrate that the electric hardness, characterizing the stability of the domain structure to external influences, is proportional to EN_{B}- (at fixed B') and EN_B (B = B', B", Ti, Zr) [1] and varies non-monotonically (passes through a maximum) with $EN_{B'B''}$.

As a measure of electric hardness [1,2], we took the uniform distortion parameter δ_t [3], since it determines the strain resulting from domain reorientations. With increasing $EN_{B''}$- and EN_B, δ_t rises (linearly in the case of EN_B), as do the mechanical quality factor (Q) and sound velocity (v_R), whereas tan δ_t, the relative permittivity of poled samples ($\varepsilon^T_{33}/\varepsilon_0$), the electromechanical coupling coefficient K_p, and the piezoelectric charge coefficient decrease, which also points to an increase in the electric hardness of the solid solutions.

The B" cations could be classified according to their electric hardness: Mn was found to be the most hard-electric, Co exhibited medium electric hardness; and Mg, Zn, and Ni were the most soft-electric. As shown recently [4], Ni is the most soft-electric cation in this series. The effect of B' and B" cations on the electric hardness of solid solutions illustrates in Table 1 [2].

Table 1. Effect of B' and B" cations on the electric hardness of solid solutions

Electric Hardness	B"	B'
Soft-electric Medium	≠ Mn, Co	Any
	Mn, Co	≠ W (except for $W_{1/2}Mn_{1/2}$)
Hard-electric	≠ Mn	Any
	Mn	≠ W (except for $W_{1/2}Mn_{1/2}$)

Experimental studies of solid solutions in more than 150 quaternary, quinary, and senary systems over wide ranges of non-PZT contents (up to 30 mol %, without predominance of the soft or hard-electric component) and Ti/Zr ratios (1.1 – 1.4) allowed the earlier derived relations to be extended to a broad spectrum of solid solutions markedly different in properties, without compositional limitations.

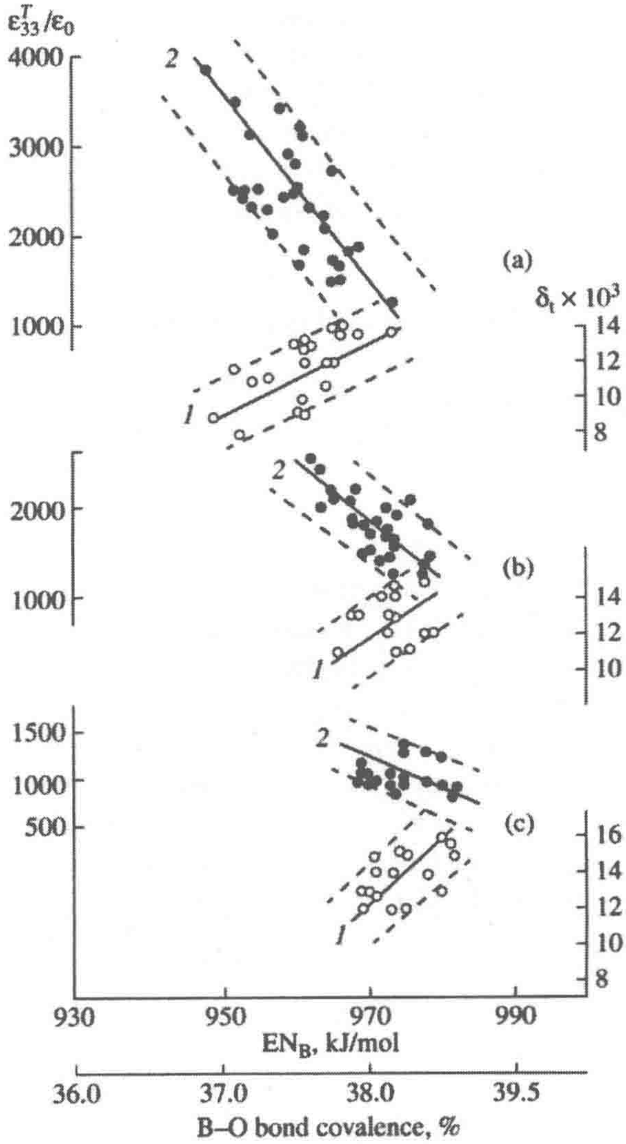

Figure 1. Plots of (*1*) δ_t and (*2*) $\varepsilon^T_{33}/\varepsilon_0$ *vs* EN_B and *B*-O bond covalence for (a) soft-electric, (b) medium hardness, and (c) hard-electric solid solutions (1).

Figure 1 displays the plots of δ_t and $\varepsilon^T_{33}/\varepsilon_0$ *vs* EN_B (solid lines) for solid solutions differing in electric hardness. The dashed lines in Figure 1 show the spread in parameters. The plots are linear and demonstrate that the electric hardness of the multicomponent solid solutions increases with increasing EN_B. A few features of these data are worth noting:

1) With increasing electric hardness, the plots shift to higher EN_B values, which provide additional evidence that the electric hardness increases with EN_B.

2) The slope of the plot of $\varepsilon^T_{33}/\varepsilon_0$ *vs* EN_B decreases and the slope of the plot of δ_t *vs* EN_B increases with increasing electric hardness, which reflects the decrease in the maximum values of $\varepsilon^T_{33}/\varepsilon_0$ and the increase in the maximum values of δ_t. Moreover, the observed variation in the slope of the plot of $\varepsilon^T_{33}/\varepsilon_0$ *vs* EN_B indicates that the sensitivity of the solid solutions to EN_B depends on their electric hardness: with increasing EN_B, the $\varepsilon^T_{33}/\varepsilon_0$ of the soft-electric solid solutions decreases more rapidly than that of the hard-electric solid solutions.
3) The spread in $\varepsilon^T_{33}/\varepsilon_0$ and δ_t decreases slightly with decreasing electric hardness (see Table 2). This is probably due to the wide diversity of $PbB'_{1-\alpha}B''_{\alpha}O_3$ constituent oxides in the soft-electric solid solutions, including both compositionally ordered antiferroelectrics (tungstates) and disordered ferroelectrics (niobates), drastically different in properties (e.g., t_c ranging from 130 °C in $PbNb_{2/3}Ni_{1/3}O_3$ to 400 °C in $PbW_{1/2}Cd_{1/2}O_3$).

Table 2. Spread in $\varepsilon^T_{33}/\varepsilon_0$ and δ_t

Electric Hardness	$\pm\Delta\ \varepsilon^T_{33}/\varepsilon_0$, %	$\pm\Delta\ \delta_t$, %
Hard-electric	29.0	16.5
Medium	29.5	17.0
Soft-electric	32.5	18.0

In hard-electric hosts, always containing manganese, the effect of some constituent oxides is reduced by the presence of Mn^{3+}, a Jahn-Teller active cation. Its contribution seems to play a key role in determining the ionic displacements and resulting dipole moment, which increases the electric hardness of such systems.

Figure 2 shows the plots of K_p and d_{31} *vs* EN_B and *B*-O bond covalence for solid solutions differing in electric hardness. These plots have a number of features in common with the plots in Figure 1: (i) they are also linear and shift to higher values of EN_B with increasing electric hardness; (ii) the largest spread in K_p and d_{31} is observed for soft-electric solid solutions; (iii) the slope of the plot d_{31} *vs* EN_B decreases and the slope of the plot of K_p *vs* EN_B increases with increasing electric hardness. At the same time, $\varepsilon^T_{33}/\varepsilon_0$ and d_{31} vary similarly, whereas δ_t and K_p vary in opposite ways. This can be accounted for using the relations [1,2,5]:

$$d_{31} \sim \varepsilon^T_{33} P_r;\ K_p \sim (\varepsilon^T_{33})^{1/2} P_r, \qquad (2)$$

where P_r is the residual reorientational polarization in the piezoelectric ceramic.

These relations follow from thermodynamic relationships [6] and experimental data [7]. The similarity in the variations of $\varepsilon^T_{33}/\varepsilon_0$ and d_{31} is due to the strong dependence of d_{31} on $\varepsilon^T_{33}/\varepsilon_0$, whereas K_p is a stronger function of P_r. Since the P_r of tetragonal solid solutions decreases with increasing 5 [1,2,5] (on account of the associated decrease in the degree of domain reorientation [1]), so does K_p, as demonstrated in Figures 1 and 2.

In contrast to the data in Figures 1 and 2, the tan δ_t *vs* EN_B data for solid solutions differing in electric hardness can be represented by a single plot (Figure 3); the same refers to the v_R *vs* EN_B data. Both tan δ and v_R vary linearly with EN_B. With increasing EN_B, tan δ decreases and v_R increases, indicating an increase in electric hardness. In both plots, the data