

Abstracts

Joint session 1: Dielectrics and Electrostatics I

Influence of electrode spacing on a symmetrical washer-type electrohydrodynamic conduction pump

M Daaboul

University of Balamand, Lebanon

Electrohydrodynamic (EHD) pumping is an attractive method that can be applied to drive a fluid in mass transport and heat transfer enhancement applications, and more particularly for microfluidic applications. EHD pumps are very interesting because they do not require moving parts. The electric energy is directly converted into kinetic energy via the Coulomb force. Three modes can generate this fluid motion: conduction, induction, and injection. This paper presents the experimental results of a parametric investigation on EHD conduction pumps with washer-type electrode geometry. Only symmetrical electrode configuration is investigated in order to highlight the influence of the positive/negative charge mobility ratio. The working fluid is a dielectric liquid (HFE-7100). The pumping mechanism is examined with several washer-type electrode geometries and different types of spacers. For each configuration, pressure and current time variations are recorded and compared.

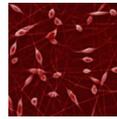
Time resolved measurement of dielectric particles velocity in standing wave electric conveyor using PTV technique

N Zouzou

Université de Poitiers, France

An efficient way to manipulate particles on a surface is by applying a multiphase voltage on a series of parallel electrodes creating standing or traveling wave electric field [1, 2]. In this paper, we study the motion of micrometric dielectric particles transported by standing waves electric field. One of the recent applications of this device is as an electrostatic dust-cleaning system to remove dust from solar panels [3]. The configuration of the standing wave electric conveyor (SWEC) is made of copper electrodes having width and inter-electrode spacing of 1 mm, and a thickness of 35 μm . The electrodes, engraved on an epoxy made surface, are energized by a 2-phase sine applied voltage. For each experiment, a mass of 20 mg of spherical PMMA particles are deposited in the middle of the board on a surface of 1 mm \times 20 mm. The particles movement is visualized and recorded using an experimental setup composed of a high-speed pulsed light LED illuminator coupled to a high-speed camera. The acquisition rate is set to 1000 Hz allowing a good analysis of the particle dynamics. Sequences of 1000 images are recorded for each experience, thus the total acquisition time is about 1 s. After recording the particles movement, a post-processing of the data is carried out using Particle Tracking Velocimetry (PTV) algorithms in order to extract information about the velocity and the position of each particle during its displacement.

In this work, we focus on the impact of the frequency on the particles displacement velocity. The frequency values are ranging from 5 to 500 Hz. The obtained results show that as a function of time, the particles behavior goes through three stages. In the first one, the particles are projected in the air under the influence of Coulomb's force ones the electric voltage is applied. In the second one, the particles pushed in the air over the surface fall down because of gravity after reaching a critical height. In the third stage, the particles jump and move toward both left and right directions. The standing wave of electric potential can be expressed as the sum of two waves that have the same magnitude, same frequency but propagate toward opposite directions. Increasing the frequency leads to the increase of the velocity of both waves. For low frequency values (less than 100 Hz), the displacement velocity of particles, in both sides, increases with the frequency at and the velocity of some of them reach the propagation velocity of the wave (synchronous



mode). For frequency values higher than 100 Hz, the particles cannot follow the fast propagation of the wave, as a consequence, the particles velocity decreases when the frequency increases.

Keywords: Standing wave, electric field, electric curtain, dielectric particles, particles velocity.

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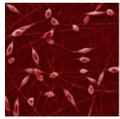
Dielectric measurements for the examination of electrostatic charging of powders

I Kiss¹, Z Á Tamus¹, T Iváncsy¹, B Németh¹, G Újfalussy² and I Berta¹

¹Budapest University of Technology and Economics, Hungary, ²Egis Pharmaceuticals PLC, Hungary

The electrostatic charge accumulation of extremely pure substances usually makes the manufacturing technology more difficult. The electrostatic behaviour of the materials can be examined by the measurement of charge decay time, but the conductivity of pure materials is very low, therefore they show very long charge decaying times. Hence, this quantity is not applicable for reliable electrostatic characterisation of pure powders. In this investigation, dielectric and charge accumulation measurements were executed on extremely pure powders. The results of leakage current and voltage response methods show significant difference between the powders according to their electrostatic behaviour. The result of dielectric measurements is confirmed by the charge accumulation measurement. The findings reveal the importance of the investigation of slow polarisation mechanisms in the charge accumulation processes of powders.

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Phased-array metasurface modeling using the MoM-GEC method

I Soltani, T Soltani and T Aguilí

National Engineering School of Tunis, Tunisia

Phased array metasurfaces are the new classes of diffractive elements through spatial phase engineering. In this article, we proposed a new design of a graphene plasmonic metasurface based on the concept of phased array antennas. Our investigation demonstrates the capability of dynamically achieve full range 360° phase modulation using graphene doping technique. This dynamic modulation is a viable solution compared to tuning the phase by varying the element length because it offers a full 2π transmission phase control.

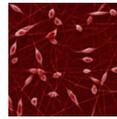
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(Invited) Electrical capacitance tomography for dielectric measurement

W Yang

University of Manchester, UK

Electrical capacitance tomography (ECT) is based on measuring capacitance from a multi-electrode sensor surrounding a subject, and reconstructing the permittivity distribution in the surrounded cross section. Obviously, ECT is a type of dielectric measurement. Various other industrial tomography techniques or modalities have been developed based on different sensing principles, such as x-ray, μ ray, optical, Laser, Terahertz, microwave, electrical resistance, electro-magnetic and ultrasonic. Among all industrial tomography modalities, ECT is the most mature and has been used for many challenging industrial applications. The internal information obtained by ECT is valuable for understanding complicated phenomena, verifying CFD models and simulation results, measurement and control of complicated processes. Compared with other tomography modalities, ECT has several advantages of no radioactive, fast response, both non-intrusive and non-invasive, withstanding high temperature and high pressure and of low-cost. Because of very small capacitance to be measured (down to 0.0001 pF) and the “soft-field” nature, ECT does present challenges in circuit design, solving the inverse problem and re-engineering. Our latest AC-based ECT system can generate online images at 300 frames per second with a signal-to-noise ratio (SNR) of 73 dB. Examples of industrial applications include the measurement of gas/oil/water flows, wet gas separation, pneumatic conveyors, cyclone separators and fluidised beds for pharmaceutical manufacturing and clean use of coal by circulating fluidised bed combustion and methanol-to-olefins conversion. During this talk, ECT will be discussed from principle to industrial applications, together with a demonstration of an AC-based ECT system.



Joint session 2: Dielectrics and Electrostatics II

Oxide based memristive devices: current status of understanding and future prospects

R Dittmann and R Waser

Forschungszentrum Juelich, Germany

Transition metal oxides exhibit a reversible, non-volatile change in electrical resistance upon electrical stimulus, a phenomenon known as resistive switching. In the simplest case resistive switching memory cells, or so called memristive devices, can be switched between a low resistance state (LRS) and a high resistance states (HRS) which can be interpreted as the logical "1" and "0", respectively. However, it is important to note that resistive switching cells often show multiple resistive states rather than only two logical states.

Based on the current knowledge, resistive switching in memristive elements based on transition metal oxides can be ascribed to electrically induced redox-processes at the oxide/electrode interface, which occur either in a spatially confined switching filament, multiple filaments or in a spatially homogeneous, area-dependent manner. In most cases, the redox-process in the metal-oxide goes along with a change in the valence state of the metal ion modifying the Schottky barrier at the oxide/electrode interface.

In this talk, we will present the current knowledge about microscopic mechanisms which drive electroforming and resistive switching in different variants of redox-based memristive elements. In particular, we will show direct experimental evidence of the redox-processes gained by in-operando spectroscopy and microscopy. Using the quantitative numbers gained from these experiments as input for existing nanoionic device simulations offers a route to less empirical and more predictive design of future memory cells. We will present detailed experimental studies and simulations of the strongly non-linear switching kinetics based on the drift-diffusion processes of oxygen vacancies in VCM-type systems. Finally, a brief overview about the current and future fields of application will be presented.

Session 3: Manufacturing of complex dielectrics and novel applications

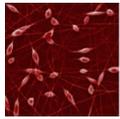
(Invited) Memristive Technologies: a viable pathway for beyond Moore electronics and AI

T Prodromakis

University of Southampton, UK

In the not so far future, electronic devices will be everywhere – embedded within our physical world and even in our bodies – empowering modern societies with unprecedented capabilities. Yet, the technological progress that brought us the mobile revolution is not any more sustainable for allowing us reaching this point. Up until now, the processing of data in electronics has relied on assemblies of vast numbers of transistors – microscopic switches that control the flow of electrical current by turning it on or off. Transistors have got smaller and smaller in order to meet the increasing demands of technology, but have nowadays reached their physical limit, with – for example – the processing chips that power smartphones containing an average of five billion transistors that are only a few atoms wide.

A novel nano-electronic technology, known as the memristor, proclaims to hold the key to a new era in electronics, being both smaller and simpler in form than transistors, low-energy, and with the ability to retain data by ‘remembering’ the amount of charge that has passed through them – akin to the behaviour of synaptic connections in the human brain. In his lecture Themis Prodromakis will present a few examples on how memristive technologies can be exploited in practical applications ranging from neuromorphic systems to charge-based computing and even enabling bioelectronics medicines.



Themis Prodromakis is Professor of Nanotechnology and Head of the Electronic Materials and Devices Research Group in the Zepler Institute, University of Southampton, UK. His work focuses on developing metal-oxide Resistive Random-Access Memory technologies and related applications and is leading an interdisciplinary team comprising 15 researchers with expertise ranging from materials process development to electron devices and circuits and systems for embedded applications. He holds a Royal Society Industry Fellowship and is a Visiting Professor at the Department of Microelectronics and Nanoelectronics at Tsinghua University, CN and Honorary Fellow at Imperial College London. He is Fellow of the IET, Fellow of the Institute of Physics, Senior Member of the IEEE and serves as the Director of the Lloyds Register Foundation International Consortium for Nanotechnology (ICoN: www.lrf-icon.com). In 2015, Prof Prodromakis established ArC Instruments Ltd, a start-up that delivers high-performance testing infrastructure for automating characterisation of novel nanodevices.

Electrocaloric effects in multilayer capacitors exceed magnetocaloric effects in prototype cooling devices

N D Mathur¹, B Nair¹, T Usui², S Crossley¹, S Kurdi¹, X Moya¹, and S Hirose²

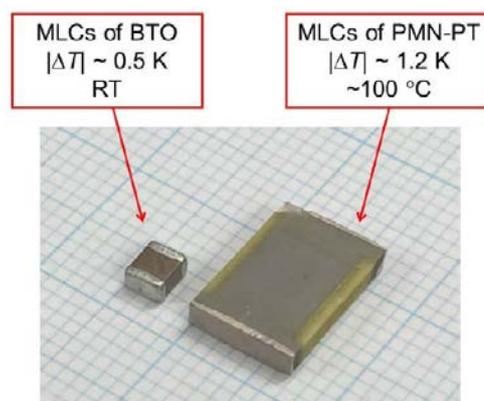
¹University of Cambridge, UK ²Murata Manufacturing Co., Japan.

There is currently growing interest in electrocaloric effects¹⁻⁴, which arise when a material displays thermal changes in response to changes of electric field, most notably near finite-temperature ferroelectric phase transitions.

I will show that bespoke multilayer capacitors (MLCs) of $\text{PbSc}_{0.5}\text{Ta}_{0.5}\text{O}_3$ (PST) display electrocaloric effects that peak at 5.5 K, and exceed 3 K over a wide range of temperatures that include room temperature.

Our peak value represents a substantial improvement over other macroscopic electrocaloric bodies, which include commercially available MLCs of BaTiO_3 (BTO) that show serendipitous electrocaloric effects of 0.5 K.

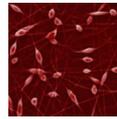
Importantly, electrocaloric effects in MLCs of PST represent an improvement over the magnetocaloric effects that are currently exploited in prototype cooling devices, where commercial-grade Gd with a substantial demagnetizing factor is addressed by 1.5 T from permanent magnets. These magnetocaloric working bodies of Gd show smaller 2.5 K changes of temperature over a smaller range of starting temperatures. We therefore suggest a straight-swap replacement, in order to improve performance and obviate the need for permanent magnets, which are bulky and expensive.



A commercially available MLC of BTO is compared with a bespoke MLC of PMN-PT [5].

Fig. 1: A commercially available MLC of BTO is compared with a bespoke MLC of PMN-PT [5]

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Dielectric meta materials using direct digital manufacturing

G Mitchell, D Sousa, S Abdulghani, and P P Faria

Centre for Rapid and Sustainable Product Development, Portugal

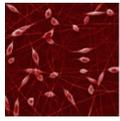
Direct Digital Manufacturing is a family of emerging family of technologies which enable a complex part to be produced directly from a digital definition without the need for complex tooling or moulds. Technologies such as 3D printing, stereolithography and selective laser melting are some of the technologies which form the Direct Digital Manufacturing Family [1].

Metamaterials are materials engineered to have a property that is not found in naturally occurring materials. They are composed of assemblies of multiple elements fashioned from materials such as metals or plastics. These materials are usually arranged in a repetitive pattern, at scales that are smaller than the phenomena they influence. Metamaterials derive their novel properties, not from the properties of the base materials, but from their newly designed structures

Much of the attention on the development of metal materials in the dielectric area has focused on generating materials with negative permittivity especially in the visible spectra as a well as microwave and terahertz regions [2,3].

This paper address how we can exploit existing direct digital manufacturing to produce objects with a repetitive structure with interesting target properties. We focus on the use of fused deposition modelling technology as this is widely used to demonstrate how to prepare meta materials. The application of such metal materials is widely based and depends on the dielectric properties achieved. In this work we review the technical limitations to using direct digital manufacturing and identify the range of properties that are possible and consider the applications which be able to exploit these new materials including the electrical simulation of scaffolds in tissue engineering.

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Session 4: Microstructure and chemistry of composite dielectrics session I

(Invited) Microstructure and chemistry in dielectric based ceramics and composites: Unveiling the potential of low temperature processing approaches

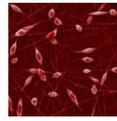
C Elissalde¹, U C Chung¹, G Philippot¹, S Basov^{1,2}, T H de Beauvoir¹, A Ndayishimiye¹, M I Rua Taborda³, A Artemenko¹, L Piraux², F Mauvy¹, S Fourcade¹, D Michau¹, C Aymonier¹, H Debéda³, C Estournès⁴, G Goglio¹, M Josse¹, and M Maglione¹.

¹University de Bordeaux, France ²University Catholique de Louvain, Belgium ³University de Bordeaux, France

⁴University de Toulouse, France

Lowering sintering temperature is a major challenge when processing refractory materials such as dielectric oxides that are conventionally sintered at temperatures between 1100°C-1400°C for several hours. High-temperature processing leads to high concentration of point defects and poor control of microstructure that both critically affect the properties of dielectric ceramics. In addition, there is a timely convergence between low temperature processes and the actual expectations of sustainability in the field of electronic components by reduction of energy consumption. The goal is to obtain high densities, chemical homogeneity, optimal microstructure and good quality of interfaces (grain boundaries, multi-materials) at low temperatures (400-900°C) while diversifying the classes of dielectric materials. Recently, significant progress have been obtained with the development of cold sintering process whose efficiency was demonstrated on an impressive number of materials.¹ In this context, our approaches rely on the use of specific low temperature processing and sintering methods (electrodeposition, soft chemistry routes, Spark Plasma Sintering in specific conditions and solvohydrothermal sintering) providing new routes to optimize and to explore dielectric based materials. Illustrations of our recent works on ceramics and composites (BST, BST/CoFe2O4, PZT, MnSO4, SiO2, ZrO2) covering a broad range of applications from capacitors, multiferroics to energy harvesters, will be given.²⁻⁶

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(Invited) Structural details of A-site substitution in LLTO perovskites. The importance of the amount of vacancies and its distribution on transport properties

J Sanz¹ and A Varez²

¹Instituto Ciencia Materiales de Madrid ICMM, Spain, ²Univ. Carlos III de Madrid, Spain

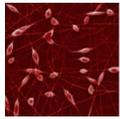
Lithium lanthanum titanate (LLTO) is one of the most promising solid electrolytes for all solid state batteries owing to its high ionic conductivity of $\sim 1 \times 10^{-3}$ S/cm at room temperature. The unusual location of Li is the key factor to understand this high Li conductivity. In particular, the amount of A-site vacancies and its distribution determine the long-range conductivity and the dimensionality of Li diffusion.

In order to understand better the ionic conductivity mechanism in these perovskites we analyzed three Sr-doped Lanthanum Lithium titanates (LLTO) series, in terms of number of vacancies and its distribution. In particular we studied the following series: (i) $\text{Li}_x\text{Sr}_x\text{La}_{2/3-x}\text{TiO}_3$ ($0.04 \leq x \leq 0.33$); (ii) $\text{Li}_{1/2-x}\text{Sr}_{2x}\text{La}_{1/2-x}\text{TiO}_3$ ($0 \leq x \leq 0.5$); (iii) $\text{Li}_{1/2-2x}\text{Sr}_x\text{La}_{1/2}\text{TiO}_3$ ($0 \leq x \leq 0.25$). In the first series, A-site are cation deficient and the amount of nominal vacancies decreases, but the effective (or total) vacancies (given by expression $n_t = \text{Li} + \square_A$) remains constant. In the second series, the amount of nominal vacancies does not change but decreases the total amount of vacancies, n_t . Finally in the third series, nominal vacancies increases as Sr content (x) increases while the n_t present a moderate decreasing ($n_t = 1/2 - x$).

The structural study of the different series indicates that slight changes in the symmetry of the unit cell do not significantly affect the transport properties. The key factor in these fast ion conductors is related to the amount of vacancies and its distribution. So the first series, showed a change of symmetry from orthorhombic to tetragonal when the lithium and strontium content increased above $x=0.08$ and from tetragonal to cubic above $x=0.16$. The Rietveld analysis of orthorhombic samples showed a preference of La ions for $z/c = 0.5$ planes, but Sr ions do not display any preference for both types of planes. Structural features observed in this series are mainly associated with the cation vacancy ordering along the c -axis, which disappear gradually when the Li content increases. In $\text{Li}_{1/2-x}\text{Sr}_{2x}\text{La}_{1/2-x}\text{TiO}_3$ ($0 \leq x \leq 0.5$) and $\text{Li}_{1/2-2x}\text{Sr}_x\text{La}_{1/2}\text{TiO}_3$ ($0 \leq x \leq 0.25$) series, there are not changes of symmetry (cubic) along the series. Rietveld analyses showed the presence of cation disorder in A-sites.

Regarding to the transport properties, it was demonstrated that long range Li motion depend on total amount of A-site vacancies, $n_t = \text{Li} + \square$. In the first series n_t remains almost constant, and d.c. conductivity do not change considerably along the series. In the case of the second series without nominal vacancies, the substitution of Li by Sr decreases the amount of n_t vacancies, reducing drastically the Li conductivity when approaches the percolation threshold of vacancies in the perovskite conduction network. Similar behavior (drop of conductivity) was found in the third series, where nominal vacancies increase but total vacancies decrease with Sr content.

Cation disordering changes slightly the percolation threshold in the first series but does not affect it in the second series. This work illustrates the important role played by vacancies in transport properties of perovskites.



Dielectrics 2019

BaTiO₃-Bi(Zn_{1/2}Ti_{1/2})O₃ relaxor materials: role of non-stoichiometry

D Cann¹, N Kumar², P Mardilovich¹, E A Patterson³, E Gorzkowski³, I Love¹, J Tucker¹, and S R Reese¹

¹Oregon State University, USA, ²The University of New South Wales, Australia, ³Naval Research Laboratory, USA

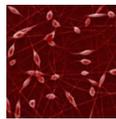
New dielectric materials designed for SiC and GaN-based power electronics technologies are needed because existing ceramic capacitor technologies are not compatible with extreme environments, especially for temperatures higher than 200 °C. In recent years, several research groups have developed new Pb-free relaxor materials that exhibit promising dielectric properties. This presentation will focus on the solid solution BaTiO₃-Bi(Zn_{1/2}Ti_{1/2})O₃ (BT-BZT) that has been shown to have a temperature independent relative permittivity, high electrical resistivity, and a relative permittivity above 1000 at fields in excess of 100 kV/cm. Structural data on these compositions show a smooth transition between tetragonal symmetry to cubic symmetry over this same range in composition. The relaxor characteristics seen in this solid solution and many similar compounds are likely due to the chemical disorder introduced through the addition of Bi³⁺ onto the A-sublattice and Zn²⁺ onto the B-sublattice of the perovskite structure. The temperature dependence can be manipulated through the addition of other compounds such as BiScO₃, BiInO₃ and NaNbO₃, for example. Furthermore, the addition of BiM O₃ to p-type BT is accompanied by significant improvement in insulation properties, often leading to an n-type behavior in BT-BiM O₃ ceramics. It will be shown that donor doping can originate primarily due to two factors—the presence of secondary phases which modifies the stoichiometry of the perovskite phase to become rich in high valence cations, and an increased oxygen vacancy concentration. It was proposed that these defects had an effect of shifting the conductivity minima to higher p O₂ values in the Kröger-Vink diagram, as a means to explain both the higher resistivity values and shift to n-type behavior while still showing a near-intrinsic activation energy. In conclusion, with an understanding of the underlying defect equilibria, BT-BZT relaxor dielectrics show promising dielectric properties with a high permittivity over broad temperature ranges which is of interest for high performance capacitor applications.

(Invited) Microstructural Imaging of grains and micron size defects of metallic structures using Quantum Well Hall Effect (QWHE) sensors

M Missous

University of Manchester, UK

Microstructural analysis of steel and related materials and imaging of magnetic domains and grains is of great importance in a range of industries. Bulk classical inspection techniques do not have the resolution (both spatial and magnetic) to extract the wealth of data available and instead rely on expensive and time consuming destructive techniques such as SEM or TEM of sectioned samples. We address these issues by using advanced Quantum Well Hall Effect (QWHE) sensors as novel solution enabling a new paradigm in high resolution microstructural analysis of materials non destructively, and without the need for any surface preparation or sectioning.



Session 5: Interpretation of dielectrics responses

(Invited) The riddle of colossal permittivity

S Hepplestone

University of Exeter, UK

Materials that exhibit colossal permittivity could lead to a wealth of new applications including a new form of high power density energy storage, sensors, electronic capacitors and a vital potential role in the global transmission of electricity across power grids. This is due to their exceptionally high dielectric response in the low frequency range with permittivities of greater than 10^3 . The most widely investigated of these materials is the archetype of colossal permittivity, $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$ (CCTO), which has shown a wide range of reported values (from 1,200 to 300,000). Here, we perform first principle calculations exploring the atomic structure of the interface and hence present a microscopic explanation for colossal permittivity in CCTO and offer it as a description for all colossal permittivity materials. In our results, we show that the high permittivity is a result of the formation of dilute metallic interfaces within the material. We discuss strategies for improving Colossal permittivity and show how the introduction of TiO_2 improves dielectric loss, but dramatically reduces the potential permittivity. The description for the origin of colossal permittivity outlined here should help guide growth techniques to optimise the colossal permittivity materials to their fullest potential.

(Invited) Anomalous dielectric relaxation in the context of the Debye model of escape of dipoles over a potential barrier

W T Coffey¹, D J Byrne^{1,2,5}, Yu P Kalmykov³, and S V Titov^{1,4}

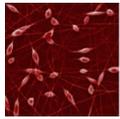
¹Trinity College, Ireland, ²Electricity Supply Board, Ireland, ³Université de Perpignan Via Domitia, France, ⁴Kotel'nikov Institute of Radio Engineering and Electronics of the Russian Academy of Sciences, Russian Federation

Anomalous dielectric relaxation in a double-well potential is reviewed showing that it explains the dielectric spectrum of CH₂Cl₂ decalin glass.

Acknowledgements

D J Byrne acknowledges Science Foundation Ireland for financial support. This publication has emanated from research conducted with the financial support of Science Foundation Ireland under Grant number 17/IFB/5420.

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(Invited) A modified Maxwell Garnett model: hysteresis in phase change materials

N G Green, X Fang

University of Southampton, UK

The dielectric properties of materials undergoing phase transitions are key to a number of modern and developing technologies, particularly when hysteresis is demonstrated in the response. This paper presents a modified Maxwell Garnett model for analysing electromagnetic hysteresis, using an asymmetric effective medium approximation to describe intermediate states in the phase change, establishing a link between effective medium and hysteresis analysis. The model has few input parameters and provides a phenomenological approach to describing electromagnetic hysteresis in various phase change materials.

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(Invited) Finite element modelling: studying the effect of microstructures in dielectric materials

J Dean

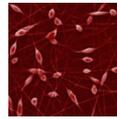
University of Sheffield, UK

Dielectric materials exhibit extremely rich and diverse physics that leads to a wide range of applications including transport, communication networks health and energy. To optimise the performance of devices based on these materials is difficult as it requires the knowledge of designing, building and testing across all length-scales.

The understanding of microstructure/property relations is central to any industrial application of dielectric materials for devices since, by knowing which microstructure features are beneficial and which are detrimental, processing techniques can be tailored and modified to exploit key criteria.

The combination of techniques such as impedance spectroscopy and microscopy are widely used to probe such features experimentally, but it is difficult to obtain systematic studies on certain features such as core/shell ratio, porosity etc.

This talk will show how experimental data (both from microscopy and measured electrical data) can be used to set-up finite element simulations and allow us to study key features for applications such as multi-layered ceramic capacitors. The systems can then be run to predict an impedance response along with the electrical



microstructure of the system (current density, electric field etc.). This data can then provide a clearer picture of the effect of the microstructure/property relations in dielectric materials.

(Invited) Analytical formula for the linear complex permittivity of isotropic polar fluids

P Déjardin, Y Cornaton

Université de Perpignan Via Domitia, France

Debye's approach of modelling dielectric properties of polar liquids [1] as developed by Kirkwood [2] and Fröhlich [3] is a useful method of calculating the static linear permittivity of polar fluids. Here, an analytical formula for polar fluids is obtained for this permittivity as a function of temperature, density and molecular dipole moment. In order to accomplish this, the Kirkwood correlation factor is calculated analytically from the averaged Dean-Kawasaki equation as given by Cugliandolo *et al.* [4] using a closure of virial type. The same closure is used to derive an analytical formula for the linear complex permittivity, where it is demonstrated that a thermally activated time scale due to intermolecular interactions is obtained when the Kirkwood correlation factor is larger than unity. In the opposite situation where the Kirkwood correlation factor is lower than unity, no thermally activated process occurs and the complex permittivity is essentially given by a Debye spectrum with amplitude modified by the intermolecular interactions [5]. The results compare reasonably well with experimental data of some simple polar fluids in their liquid phase [6]. Furthermore, favorable comparison with various numerical simulations of the static dielectric permittivity as a function of temperature is also obtained for liquid water and liquid methanol as examples.

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Session 6: Measurement of dielectrics responses

Measurements on low-loss dielectrics in the frequency range 1 to 70 MHz by using a vector network analyser

A P Gregory and G J Hill

National Physical Laboratory, UK

Resonance measurements on a dielectric test-set [1] in series or parallel with a coil can be used to determine the permittivity (ϵ') and dielectric loss ($\tan \delta$) of materials in the RF range. Very low loss materials can be measured with a resolution of < 10 microradians [2]. This is appreciably better than that obtainable by impedance-based methods. The Lynch (equivalent thickness) method [3] is used.

In the 1970s much work went into developing Q-meter based systems, based on series resonance, for measuring low loss insulating materials (e.g. LDPE) used in coaxial submarine telephone cables. There is continuing interest in the measurement of the loss of very low loss materials that are used in the electronics industry, and as windows in high-power RF systems.

We outline the development of a new system that replaces the obsolete Q-meter with a Vector Network Analyser (VNA), using parallel resonance (Fig. 1). Measurements made on the old and new systems are compared for HDPE, Macor and alumina specimens, and show good agreement.

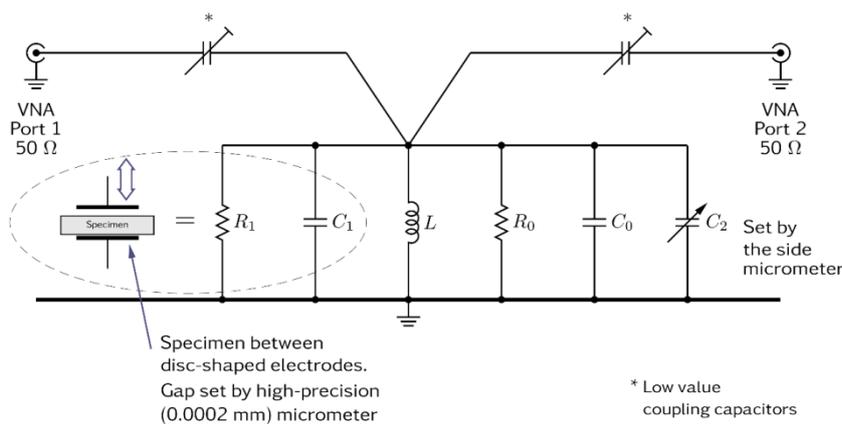
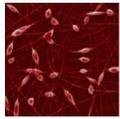


Fig. 1: Equivalent circuit for measurement using a VNA by transmission. This uses a parallel LC resonant circuit. C_0 represents stray capacitance. R_0 represents conductor loss, e.g. in the coil.

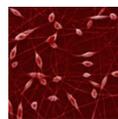
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(Invited) Distortions in the structures of the twist bend nematic phase of a bent-core liquid crystal by electric field using dielectric spectroscopy

J Viji

University of Dublin, Ireland

The dielectric spectra of the twist bend nematic phase (NTB) of an achiral asymmetric bent-core liquid crystalline compound are studied for determining the various relaxation modes in the frequency range 1 Hz to 100 MHz. Dielectric measurements are carried out on an asymmetric bent-core liquid crystal using a low probe field. This is superimposed on the bias field E that is varied up to $8 \text{ V}/\mu\text{m}$. Two molecular and two collective relaxation processes are observed. The orientational order parameters with respect to the local as well as the main director are determined using molecular modes. This is used to find the heliconical angle. The results show that the order parameter with reference to the coarse-grained director reverses its trend from increasing to decreasing at temperatures few degrees above the NTB to N transition. The collective relaxation modes are assigned to (a) the distortions of local director by the electric field at a frequency of $\sim 100 \text{ kHz}$ while the periodic helical structure remains unaltered; this mode is attributed to flexoelectricity (b) to the changes in the periodic structure that arise from a coupling of the dielectric anisotropy with the electric field at the lowest frequency of investigation in the range of 1 Hz–10 kHz. Frequency of the higher frequency collective mode ($\sim 100 \text{ kHz}$) depends primarily on the heliconical angle and has anomalous softening like behaviour at the N-NTB transition. The lowest frequency mode is studied under the bias field E ; the modulus of the wave vector gradually vanishes on increasing E (except for an initial behavior, $E^2 < 0.1 \text{ V}^2/\mu\text{m}^2$, which is just the opposite). The transition from the twist bend to the splay bend structure is observed by a sudden drop in the frequency of this mode. This is followed by a linear decrease in its frequency by increasing E . The results agree with the predictions made from currently proposed models for a periodically distorted NTB phase.



New functionalities from ferroelectric domain walls

M Gregg

Queen's University Belfast, UK

Over the last decade, it has become clear that domain walls in ferroelectrics and multiferroics can have functional properties that are very different from those of the domains that they surround. Enhanced domain wall conductivity is often observed, usually as a consequence of charge accumulation or band-structure changes at walls across which there are discontinuities in polarisation. Conducting walls have been considered for applications in novel agile nano circuitry, making and breaking electrical connections as they can be injected, moved and annihilated by applied fields. However, they can be potentially much more exciting in making completely new forms of devices. In this talk we will discuss how domain walls can be used to create new kinds of memristors, how 1D p-n junctions can be made inside domain walls and how the unexpected movement of specific charged walls in boracite systems could create a new avenue in negative capacitance.

Session 7: Interpretation of dielectrics responses session II

(Invited) Understanding and controlling the dielectric response of porous materials

M Ryder

Oak Ridge National Laboratory, USA

Highly porous materials such as metal-organic frameworks (MOFs), covalent organic frameworks (COFs), and hydrogen-bonded organic frameworks (HOFs) have recently been shown to have promising electronic and dielectric properties.[1-3] In this talk I will discuss a diverse range of porous materials, primarily MOFs, to rationalize how the different building blocks that form the structure can affect the electronic properties and dielectric response. The results, obtained using density functional theory (DFT) calculations, study the contribution from the metals involved, the organic linkers and the symmetry and topology of the framework. The work confirms that the electronic band gap is primarily due to the electronic levels of the organic linkers and that tuning the band gap can be achieved either by linker functionalization or by increasing the aromaticity. The relevance of simple structure-property relationships for different families of isorecticular MOFs using Hammett sigma constants is also highlighted. It is also shown that the polarizability of the framework can be tuned comparably to the band gap. However, the expected low static dielectric constant is less influenced by the composition of the MOF and can be modified by acting on the crystal structure.

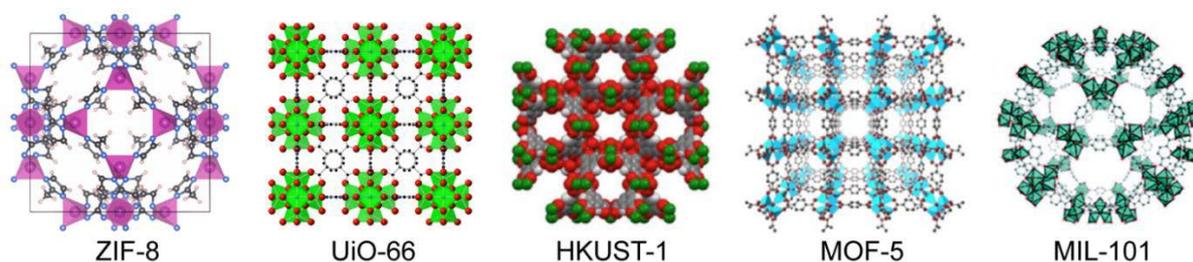
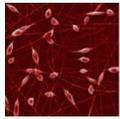


Fig. 1: MOF materials showing long-range nanoscale porosity and framework topologies.

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(Invited) Relaxor behaviour and disorder in tetragonal tungsten bronzes

F Morrison

University of St Andrews, UK

The majority of studies of ferroelectric materials focus on those which adopt the perovskite (ABO_3) structure, the closely-related tetragonal tungsten bronze (TTB) structure, have been much less studied. TTBs, described by the general formula $A_1A_2A_4B_1B_2B_3C_4O^{30}$, offer a similarly wide degree of compositional modification to perovskites, but in addition also offer degrees of freedom due to non-degenerate A- and B- crystallographic sites and also a high degree of non-stoichiometry. This mixture of structural and compositional flexibility results in equally complex crystallography with a number of possible distortions from the centrosymmetric $P4/mbm$ aristotype structure. These include simple loss of inversion symmetry, orthorhombic and monoclinic distortions involving very large unit cells, and also incommensurately modulated structures. This leads to a range of electrical properties including dipole glass, relaxor ferroelectric and 'normal' ferroelectric behaviour – often in the same composition as a function of temperature.

Again, in contrast to perovskites our understanding of composition-stoichiometry-property relationships is poor by comparison. The role of composition and factors affecting structural disorder which controls relaxor behaviour in TTBs will be presented. I will discuss a recent examples of TTB materials which exhibits both relaxor ferroelectric and 'normal; ferroelectric behaviour where long-range, "average" structural data, as determined by "conventional" synchrotron and neutron powder diffraction, was insufficient to rationalise the macroscopic electrical properties and a combination of selected area electron diffraction and total neutron scattering was required to probe an order-disorder crossover in these materials.

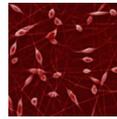
Potentialities of flexoelectric effect in soft polymer films for electromechanical applications

B Guiffard¹, M Saadeh^{1,2}, P Frère², R Seveno¹, M El-Gibari¹, T Sghaier¹, V I Merupo^{1,3}, and A Kassiba³

¹Université de Nantes, France, ²Université d'Angers, France, ³Université du Maine, France

Among the transduction mechanisms of interest for sensing and/or actuation applications at nano/micro scale, the piezoelectric effect has been widely exploited owing to the solid state nature of piezoelectrics, the large ability of specific classes of materials for the mechanical-to-electrical energy conversion and easy integration. However, every piezoelectric (also generally ferroelectric) presents well-known intrinsic drawbacks such as required poling step and related aging. In contrast, uniquely flexoelectric materials do not suffer from these disadvantages because flexoelectricity, a universal effect in all dielectric solids defined as the electrical polarization induced by a strain gradient, does not imply preliminary electric field-induced macroscopic polarization. Besides, strain gradient may be easily obtained by bending plate or cantilever-shaped structure and in this case it is nothing but the local curvature of the flexible system. Thus, as strain gradient (curvature) inversely scales with both elastic stiffness and thickness, this study will focus on the evaluation of the potentialities of flexoelectric effect in soft polymer films for electromechanical applications, with an emphasis on the thickness influence. In this way, analytical results combined to experimentally obtained effective flexoelectric coefficients for some typical polymer classes may provide guidelines for the development of soft and low frequency flexoelectric mechanical transducers.

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Combination of voltage response method with non-contact electrostatic voltage measurement to determine the dielectric response of insulating materials

Z Á Tamus

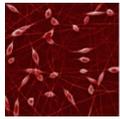
Budapest University of Technology & Economics, Hungary

The paper introduces the development of a new voltage response measurement method to determine the dielectric response function of insulating materials. This new method is based on a non-contact electrostatic voltage follower. In this technique, the voltage of the measuring probe follows the surface potential. The probe voltage is connected to the guarding electrode and helps to increase the impedance of the equipment. The paper introduces the built-up of the equipment and the relationship between the slopes of decay and return voltages and the dielectric response function.

Acknowledgments

Project no. 123672 has been implemented with the support provided from the National Research, Development and Innovation Fund of Hungary, financed under the KNN 16 funding scheme.

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Charged interfaces in ferroelectric materials

M Maglione

University of Bordeaux, France

As for any semi-conductor material, free charges maybe localised at interfaces in ferroelectric materials. This localization may have large and hopefully controlled effect on the functionality of these materials. I will review three such cases:

-electrodes interfaces in BaTiO₃ single crystals where free charges maybe generated by controlled reduction thus increasing the apparent dielectric permittivity at high temperature (T>600K) [1]

-domain wall within the ferroelectric phase of BaTiO₃ where Fe-related free electronic charges were shown to induce an artificial magneto-capacitance effect [2]

-in KTiOPO₄ single crystals, it is the localisation of free K⁺ ions at the surface and at domain walls which raises the Second harmonic Generation coefficients

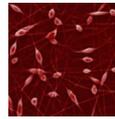
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DOI:10.1364/OME.6.000137

Correlation between structure with dielectric and ferroelectric properties for the lead-free electro-ceramics based on Ba_{1-x}Ca_xTi_{0.9}Zr_{0.1}O₃

G Herrera-Pérez¹, A Reyes-Montero², J R Plaisier³, A Reyes-Rojas⁴, and L. E. Fuentes-Cobas¹

¹ Centro de Investigación en Materiales Avanzados S. C., Mexico ² Universidad Nacional Autónoma de México, Mexico ³ Elettra-Sincrotrone Trieste S.C.p.A, Italy

The scientific community and the electro-ceramic industry are interesting on replacing leadbased materials due to their toxic effects and unfavourable environment impact. The motivation of the present work is to establish the relationship between the crystal structure, microstructure with the piezo-ferroelectric properties emphasizing in the dielectric properties such as the dielectric loss tangent ($\tan \delta$), the permittivity (ϵ), Curie-Weiss temperature (T_{CW}) and the diffusion coefficient (γ). The compositions proposed in this work are based on the Ba_{1-x}Ca_xTi_{0.9}Zr_{0.1}O₃ stoichiometry (where x = 0, 0.1 and 0.15). These compositions show a single perovskite structure and they are located near the so called morphotropic phase boundary (MPB): coexistence of ferroelectric phases. High-resolution X-ray diffraction (HR-XRD) patterns were collected at the beamline 7.1 MCX Elettra Sincrotrone in order to monitor possible changes of electronic structure profile due to the local distortions as Ca²⁺ increases. HR-XRD results suggest the coexistence of tetragonal and rhombohedral phases. Figure 1(a) shows a scanning electron micrograph that reveals the presence of two morphologies with an abnormal grain growth. The piezoelectric coefficient d_{33} and ferroelectric properties (panel b) such as remanent polarization P_r and coercive field E_c complement the electrical characterization



for these compositions. The dielectric analysis performed on the curves showed at panel c and d, indicates a relaxor behaviour with a Curie temperature in a range of 100-120 °C (panel e and f).

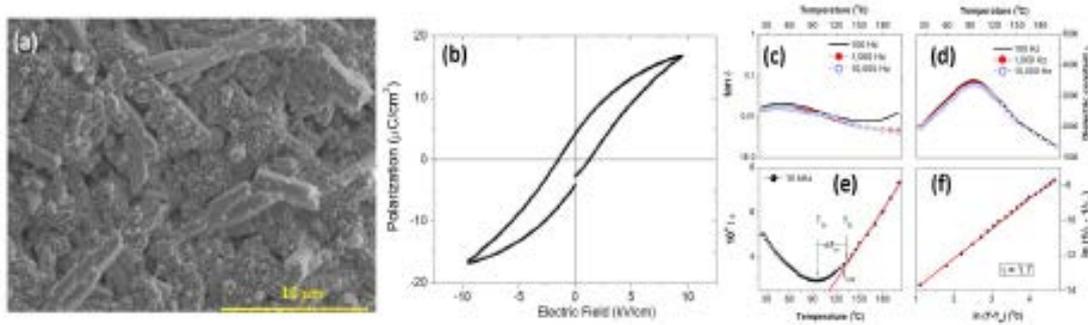
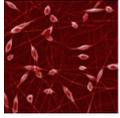


Figure 1. (a) Scanning electron micrograph reveals a two different grain sizes and shapes. (b) Ferroelectric hysteresis loops for BCZT $x = 0.1$ ceramic. (c) Dielectric loss tangent. (d) Dielectric constant. (e) Inverse dielectric permittivity ($1000/\epsilon'$) as a function of temperature at 10 kHz. (f) Plot of $\log(1/\epsilon - 1/\epsilon_m)$ versus $\log(T - T_m)$, dots: experimental data, solid line: fitting.



Poster programme

(P1) Monitoring permittivity of cable insulation polymers using inter-digital capacitive sensors

N Bowler

Iowa State University, USA

The development and performance assessment of inter-digital capacitive sensors for in situ monitoring or testing of cable insulation materials will be described. Degradation of cable insulation materials can lead to electrical breakdown or arcing between conductors that may result in loss of control function or fire. Loss of control function is especially dangerous in the context of aircraft flight control, or in the operation of a nuclear reactor, for example. Time- and frequency-domain reflectometry are useful methods for locating faults in the electrical conductor of a cable, but are less sensitive to degradation-related changes in the insulation material that manifest as a change in its permittivity. Inter-digital capacitive sensors can be applied to a particular location on the surface of a non-shielded cable and a measurement of capacitance made, from which the condition of the cable insulation material may be inferred.

By judicious design of the sensor, sequential measurements of capacitance can be made that are sensitive to either near-surface or sub-surface material changes. A finite-element model (COMSOL®) is then applied that accounts for the sensor and cable geometry, enabling permittivity to be inferred from the capacitance measurements. In this way, the signal due to material changes in the all-important insulation layer, that directly contacts the electrical conductor, can be separated from signal changes due to material changes in the near-surface cable jacket.

As an example of the method, the relative permittivity of ethylene propylene rubber (EPR) insulation material is inferred from capacitance measured using an inter-digital capacitive sensor positioned on the surface of a single-conductor cable jacketed with chlorinated polyethylene, that had been aged thermally at 140 °C for 0, 7, 14, 21, 28, and 35 days. It is shown that the relative permittivity of the EPR insulation material inferred from these measurements correlates closely with relative permittivity measured on planar control samples aged under similar conditions.

It is concluded that inter-digital capacitive sensors are promising for localized monitoring of cable insulation degradation by observing changes in the permittivity of the insulating material.

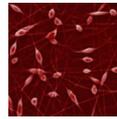
(P2) Bond- graph input-state-output port-Hamiltonian formulation of memristive networks for emulation of Josephson junction circuits

S Hadjiloucas

University of Reading, UK

State space transformations from a bond graph representation of the Josephson junction are developed, and then an analysis that links the associated inputs and outputs in the junction to the nonlinear characteristics of the memristive element is provided. A bond graph Input-State-Output Port-Hamiltonian formulation of memristive networks for the analysis of Josephson junction circuits is presented.

The interconnection between the storing energy elements (inductors and capacitors) and dissipative energy elements (resistors and memristors) that enable the energy exchange across ports as well as the energy exchange with the environment is modeled through the assumption that they represent voltage and current sources. Using Kirchoff's current and voltage laws the network model is subsequently generated. In the Port- Hamiltonian framework the structure of power flow within the system is also incorporated, furthermore, the total energy flow reflects the circuit physical structure (connectivity) so that a Hamiltonian Function for the circuit can be generated.



In BG theory, power is the result of the product between effort $e(t)$ and flow $f(t)$. Flow and effort variables at all the ports of the network are described using the causal bond graph methodology. The causality concept is used to assign the direction of power-conjugated input-output pairs [1]. As discussed in [2], a BG general structure is composed of input/output dissipation fields (D) (with subscripts i and o respectively) that can be split into two parts: linear (I) and nonlinear memristive ones (M). The formulation also defines storage fields (C and I), source fields associated with effort and flow (S_e and S_f), and junction structures (denoted by JS) containing transformers TF and gyrators GY as shown in Fig. 1. Dissipation is seen as composed of input and output variables. The dissipation variables consist of two types of elements: linear and nonlinear. Memristive dissipative elements using the BG framework are incorporated in the general junction structure shown below.

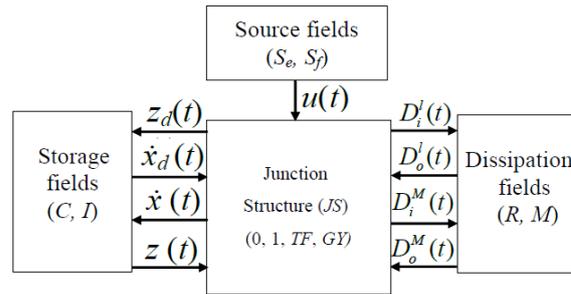


Fig. 1 Structure of a memristive causal bond graph with source, storage and dissipation fields.

A defined junction structure for systems with a memristor can be developed in state space (with states x and inputs u) using the generic expression shown below.

$$\begin{bmatrix} \dot{x}_i(t) \\ D_o^I(t) \\ D_i^M(t) \\ z_d(t) \end{bmatrix} = \begin{bmatrix} S_{11} & S_{12} & S_{13} & S_{14} & S_{15} \\ S_{21} & S_{22} & S_{23} & S_{24} & S_{25} \\ S_{31} & S_{32} & S_{33} & S_{34} & S_{35} \\ S_{41} & S_{42} & S_{43} & S_{44} & S_{45} \end{bmatrix} \begin{bmatrix} z_i(t) \\ D_o^I(t) \\ D_o^M(t) \\ u(t) \\ \dot{x}_d(t) \end{bmatrix} \quad (1)$$

The methodology has applications to the modeling of SQUIDs and other non-linear transducers and enables the formulation of input-output models of complex components embedded in non-linear networks. Examples of such models will be presented at the conference.

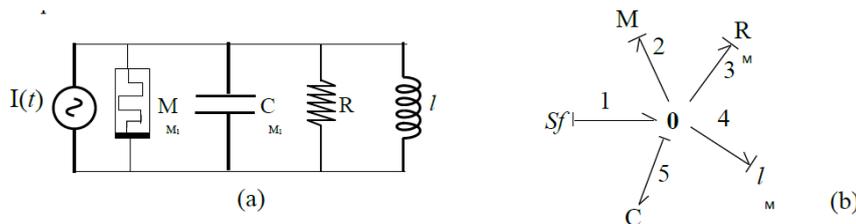
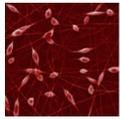


Fig. 2: (a) Josephson junction circuit model with the non-linearity emulated using a memristor. (b) The corresponding bond graph with causality marks.

[1] W. Borutzky, *Bond Graph Methodology: Development and Analysis of Multidisciplinary Dynamic System Models*, vol. 26. London, UK: Springer Science & Business Media, 2009.
 [2] G. Avalos and R. Orozco, "A procedure to linearize a class of non-linear systems modelled by bond graphs," *Math. Comput. Model. Dyn. Syst.*, vol. 21, no. 1, pp. 38-57, Jan. 2014.



(P3) A Modified Maxwell Garnett Model: Hysteresis in phase change materials

N G Green

University of Southampton, UK

Phase change materials such as chalcogenide glasses switch between two phases with greatly different properties, with critical technological applications such as rewritable optical discs and non-volatile electronic memory. The control of intermediate states in these materials is also of interest in, for example dynamically tuneable antennae, beam steering and multi-level memory. Transitions between phases often involves complicated atomic and electronic changes, a topic of significant study in condensed matter physics and material sciences. Hysteresis is often evident in phase change materials, which for thermally triggered phase changes, signifies different properties upon heating and cooling even at the same temperature. In memory applications in particular, large and controllable hysteresis is integral to their function. Analytical modelling of hysteresis is important for insight into phase transitions in materials and for, for example Preisach, Prandtl-Ishlinskii and Duhem. The choice of model, often with modification, depends on the specific material and/or properties under study, however while useful for analysis, the application to design is limited by complexity.

This paper presents an analytical model for describing electromagnetic hysteresis, based on the Maxwell Garnett effective medium approximation. The model is generic and developed for use on arbitrary phase change materials. The model uses an asymmetric effective medium approximation to describe intermediate states in the phase change, establishing a link between effective medium and hysteresis analysis. The model is simple, requires very few input parameters, and provides a phenomenological approach to describing electromagnetic hysteresis in various phase change materials. Example calculations are presented, demonstrating the effects of hysteresis and the determination of conditions in intermediate states.

(P4.) Correlation between structure with dielectric and ferroelectric properties for the lead-free electro-ceramics based on $\text{Ba}_{1-x}\text{Ca}_x\text{Ti}_{0.9}\text{Zr}_{0.1}\text{O}_3$

G Herrera-Pérez¹, A Reyes-Montero², J R Plaisier³, A. Reyes-Rojas⁴, and L. E. Fuentes-Cobas¹

¹ Centro de Investigación en Materiales Avanzados S. C., Mexico ² Universidad Nacional Autónoma de México, Mexico ³ Elettra-Sincrotrone Trieste S.C.p.A, Italy

The scientific community and the electro-ceramic industry are interesting on replacing leadbased materials due to their toxic effects and unfavourable environment impact. The motivation of the present work is to establish the relationship between the crystal structure, microstructure with the piezo-ferroelectric properties emphasizing in the dielectric properties such as the dielectric loss tangent ($\tan \delta$), the permittivity (ϵ), Curie-Weiss temperature (T_{CW}) and the diffusion coefficient (γ). The compositions proposed in this work are based on the $\text{Ba}_{1-x}\text{Ca}_x\text{Ti}_{0.9}\text{Zr}_{0.1}\text{O}_3$ stoichiometry (where $x = 0, 0.1$ and 0.15). These compositions show a single perovskite structure and they are located near the so called morphotropic phase boundary (MPB): coexistence of ferroelectric phases. High-resolution X-ray diffraction (HR-XRD) patterns were collected at the beamline 7.1 MCX Elettra Sincrotrone in order to monitor possible changes of electronic structure profile due to the local distortions as Ca^{2+} increases. HR-XRD results suggest the coexistence of tetragonal and rhombohedral phases. Figure 1(a) shows a scanning electron micrograph that reveals the presence of two morphologies with an abnormal grain growth. The piezoelectric coefficient d_{33} and ferroelectric properties (panel b) such as remanent polarization P_r and coercive field E_c complement the electrical characterization for these compositions. The dielectric analysis performed on the curves showed at panel c and d, indicates a relaxor behaviour with a Curie temperature in a range of 100-120 °C (panel e and f).

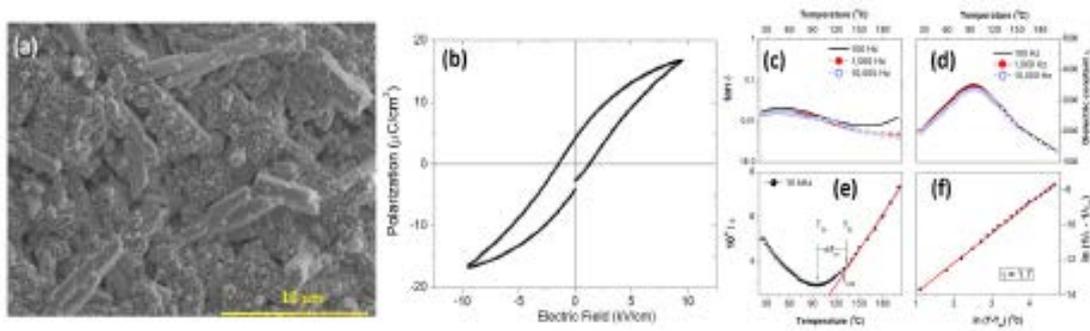
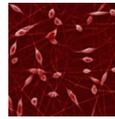


Figure 1. (a) Scanning electron micrograph reveals a two different grain sizes and shapes. (b) Ferroelectric hysteresis loops for BCZT $x = 0.1$ ceramic. (c) Dielectric loss tangent. (d) Dielectric constant. (e) Inverse dielectric permittivity ($1000/\epsilon'$) as a function of temperature at 10 kHz. (f) Plot of $\log(1/\epsilon' - 1/\epsilon_m)$ versus $\log(T - T_m)$, dots: experimental data, solid line: fitting.