Oil Saturation Effects in Lead MetaNiobate Porous Piezoceramic: Transient Material Characteristics

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Abstract — Lead metaniobate $PbNb_2O_6$ (PN) has a unique combination of high piezoelectric anisotropy near 10 (thickness-to-planar coupling coefficients), relatively low dielectric permittivity and high Curie temperature, low *Q*-factor near 20. The latter is the most intriguing PN property in the row of piezoelectric materials, and as was shown in the present research this internal high dissipation effect is directly related to the presence of silicon oil in the porous PN structure, and consequently is determined by the oil properties. To the contrary, the quality factor of PN not saturated with oil was found as high as near 400. Full sets of PN electro-mechanical constants, transient resonance and dissipation characteristics and their temperature dependences were determined under both conditions: conventionally saturated and not saturated with oil. As was experimentally shown, at higher temperatures particularly 260°C soak for several days, a transition from "with oil" to "no oil" in the PN properties takes place, and this effect is a consequence of the phase transition in oil, from liquid to solid state.

Keywords: **piezoelectric materials; piezoceramics; material properties; piezoelement; quality Q-factor; silicon oil; resonance and antiresonance frequencies; temperature stability.**

I. INTRODUCTION

Lead metaniobate $PbNb_2O_6$ (PN) is an outstanding piezoceramic [1,2] in the row of more common piezoelectric materials, like lead-zirconate-titanate (PZT), with not just compositional differences. PN is highly anisotropic with k_t/k_p near 10, it has high Curie temperature near 500°C, extremely low Q_m factor near 20, low acoustic impedance, and low dielectric constant; it is primarily used in NDT systems where low Q_m factor and clean impulse response are required [3,4]. A close alternative is a PZT piezocomposite [5], however its application temperature is highly restricted by the properties of epoxy used in the structure. There are several leading piezoceramic companies currently promoting their lead metaniobate brands for industrial applications, such as basic suppliers [4,6,7] Meggitt Sensing Systems (Pz-35, K-81&85), PiezoKinetics (PKI-100&105), SensorTech (BM-901&941), DelPiezo (DL-31,32,33), Elpa, etc.

In general, three crystalline forms of PN are known: low- and high-temperature thermodynamically stable rhombohedral and tetragonal, respectively, and ferroelectric metastable orthorhombic phase [4,8,9]. The latter can be obtained by quenching the samples rapidly to lower than Curie temperature from the sintering high temperature. Meanwhile, such a fast temperature process widely creates a predominantly open net of tiny pores [8,9], which particularly are filled with silicon oil in the necessary conventional polarization process.

Multiple GeoPhysics researches stated that acoustical wave propagation through partially or fully fluid-saturated rock materials creates complex interaction between the fluid and solid phases [10-12]. In the presence of pores, the fluid not only acts as a stiffener to the material, but also results in the flow of the fluid between regions of higher and lower pore pressure [12], creating additional damping.

When a compressional wave squeezes that medium, local pressure gradients occur as a consequence of the matrix deformation and subsequent flow of the local pore fluid. The behavior of fluid in the pore space makes the elastic moduli of the porous material frequency-dependent [12]: a) at high frequencies, the fluid in the pore structures becomes isolated, causing the material to be significantly stiffer, with reduced energy losses; b) at low frequencies, the bulk moduli and dissipation of the porous medium depend basically on the frame properties, with c) a transitional zone at median frequencies where the properties depend predominantly on the flow properties of fluid in the medium frame. Particularly at statics at least with open pores, the stiffness seems to be not dependent on the liquid saturation. In [11] the authors provided a specific link between flow under effective compressibility of porous media and acoustical properties, a goal which is a target for the piezoceramic-physics research of porous piezo structures.

When a structure vibrates in a mode with local compression and expansion, just to note, there are always some extra losses associated with the irreversible heat conduction between cool and heated areas. These losses result in additional thermoelastic (acoustic) damping which is most pronounced when the vibration frequency at a certain vibrational mode is close to the thermal relaxation frequency [13], presumably increased by the inner fluid flow effect in a porous body.

Soft lead zirconate-titanates (PZT) and lead metaniobates piezoceramics, because of their relatively high damping, are normally used to manufacture broad-band piezoelectric ultrasonic transducers, to provide high time/space resolution and measure dynamic phenomena. It is therefore necessary to use ultrasonic transducers capable of sending short signals without post-pulse oscillations. However there are a number of differences between those piezoceramic types. Soft PZTs having a moderate Q-factor near 100 are well known and tested materials, easily manufactured, characterized and widely used. On the contrary, the characterization of lead metaniobates is difficult because of at least relatively high damping and extremely low planar coupling coefficient, all under the effect of oil saturation [6,7], and a number of precautions must be considered to handle the measurements [14-16]. High damping of lead metaniobate is of special interest in that context because its damping is intrinsic to the piezoelement structure; it does not depend on the presence of an external damping circuit, while the material is relatively stiff. In the literature, low-Q near 20 and high-Q near 500 PN ceramics are described (for example [2,6]), without detailed investigation and explanation such a difference.

Three different levels of PN material characterization appear to take place: both conventional oil-saturated and without oil structures to describe the transducer extrinsic acoustical performance, and of intrinsic PN material itself (monolithic frame body of the structure) as an internal local material characterization. In the present work, a full set of the global elastic, dielectric and piezoelectric material constants were determined for no-oil and oil-saturated PN piezoceramic, and the basic electro-elastic parameters of the main PN body were estimated.

Some commercial piezoceramics have open and interconnected porosities, filled with polymers, such as conventional piezocomposites. Therefore the material is no more homogeneous. Typically the 3-3 connectivity of the pore space (like a coral structure) in the ceramic makes possibilities to fill in the ceramic by infiltrating a fluid or to drain it completely. As previous works have shown [17-20], the minor phase has a strong influence on the properties of the porous ceramic. Several theories [5,17,21,22] have been developed to explain the changes of the properties of a ceramic when the minor phase is changed, focusing on its influence on the overall transducer dynamic performance.

Basically as was concluded, the composites are not a linear combination of the properties of each different form, and interaction between both phases has to be considered taking into account the different ways in which the two phases of the porous ceramic interact.

In [23] it was shown that conventional lead metaniobate has a significantly lower dissipation (damping) at infrasonic frequencies, which may be encountered in bimorphs or other bender elements, than it does at ultrasonic frequencies used in determining the ‘book value’ of extremely low Q-factor. In general, the mechanical damping in ferroelectrics can occur as a result of interaction between stress-induced domain wall motion and defects, including dislocations and point defects such as vacancies. As was stated in [23], the damping in PN in a wide infrasonic to ultrasonic frequency range is not of the hysteresis type, so that the known mechanisms that give rise to such behavior cannot play a major role. It was concluded that the attainment of a complete understanding of the causal mechanisms for damping in PN is not clear yet. In the present work a new dissipation mechanism type with the fluid-structural dynamic interaction was proposed and experimentally analyzed for PN piezoceramic.

Moreover as reported, PN piezoceramic occasionally exhibits “forbidden” even-order mechanical resonances, particularly at the thickness resonance [18]. Unexpected “forbidden” mechanical resonances of relatively low amplitude, which approach at double the thickness mechanical resonance and the rest of even overtones, have been observed in some PN disc samples. No explanation of this effect had been found, and it was not clear if these resonances were mechanical or not. The only common characteristic in all the samples is that all of them have interconnected internal porosity.

Just notice one of the prominent anomalies in PN behavior described in the literature [24], with a thermal expansion phase transition found at low temperatures near -30ºC.

In general, the present technological and theoretical research provides a comprehensive explanation of the phenomena involved in the characterization of a porous PN piezoceramic.

A. Scope of the Present Research, for Oil and Alcohol PN Saturation.

Specimens of lead metaniobate were prepared from commercially available transducer disks [6], with fired silver electrodes. They were polarized either traditionally in “oil”, or high pressure “air” polarization was used as well for some particular experiments. Typically the commercial silicon oil is used for polarization, particularly Molykote 200-20 type, with relatively low viscosity. After polarization the samples were chemically cleaned with alcohol for near 30 min.

The estimated effective porosity of the samples is not more than 4% max. Basic characterization was made using an HP 4194A Impedance Analyzer with relatively low 0.5 V maximum voltage applied to the sample; the Q-factor was determined as an inverse relative frequency bandwidth between the frequency extremes [16] of the imaginary part B of admittance $Y = G + iB$ for resonance, and the imaginary part X of impedance $Z = R + iX$. A Berlincourt meter was used to measure d_{33} quasistatic piezocoefficient. As was previously noticed in the literature [18], in experiments and based on the Banno's model [5], when the minor phase is a fluid (not for solid fillers), the piezoelectric coefficients do not change much.

Variation of the properties of porous piezoceramic was experimentally investigated when a minor phase is put inside the pores. Three cases were analyzed: 1) “air” saturation which is used as a reference as the effect of the minor phase can be neglected in this case, 2) silicone oil saturation (low viscosity oil used for polarization) and 3) alcohol saturation (light volatile liquid).

The global piezoceramic properties were considered under influence of: 1) the properties of the major and minor phases – some volume fraction that is occupied by it, and 2) the mechanical and electrical interaction between those both phases.

The electrical impedance around radial and thickness resonances of a PN disc was measured before and after saturating it with silicon oil, or alcohol, to simulate the effect this disc could perform.

The piezoelement dimensions were chosen close to most popular in practice. The characterization of the general basic piezoelectric and mechanical parameters was made both under normal conditions (NC) and elevated temperatures.

II. GENERAL COMPARATIVE PIEZOCOMPOSITES PERFORMANCE

A. *Comparative Piezocomposite 1-3 vs. Lead MetaNiobate Resonance Characteristics*

If compare “conventional” low-Q, high anisotropy PN with traditional epoxy-filled 1-3 PZT piezocomposite, both are used to provide similar high-damping acoustical performance. As shown in Fig. 1, two corresponding representative piezoelements were taken with close resonance frequencies and electrical impedances. Resonance intensity of the radial mode relative to the thickness mode intensity is significantly less in the case of PN. Note that for a typical PZT (not composite), radial and thickness resonance intensities are near equal.

Notice some differences in behavior of the two piezoelement structures. As mentioned, two basic conditions in PN were investigated here: “original” without oil, and “conventional” saturated with silicon oil, traditionally used in the PN polarization process. In the 1-3 PZT piezocomposite filled with a relatively soft epoxy filler, compared to an original homogenous piezoceramic plate, the fundamental resonance frequency is less and basically determined by the stiffness $1/S_{33}^{E}$ (Young’s module) of the ceramic mini-rods of the 1-3 structure, while in the homogeneous plate by the C_{33}^{E} stiffness. In PN the situation is opposite – for the conventional PN saturated with oil, its fundamental resonance frequency is higher and roughly determined by the PN intrinsic stiffness C_{33}^{E} , while for the original PN without oil inside the plate it is determined by the PN intrinsic Young’s module $1/S_{33}^{E}$.

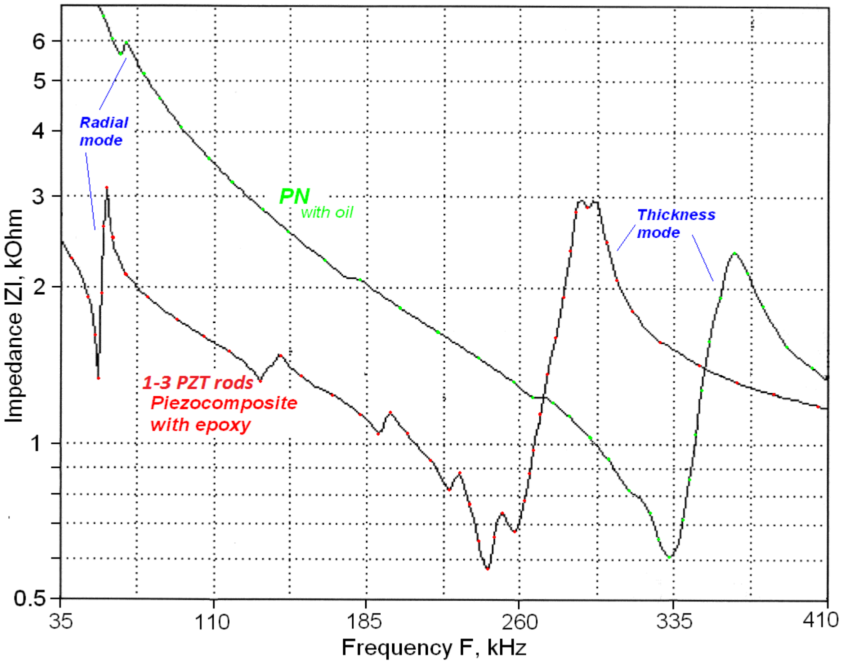


Fig. 1. Comparative electrical impedances IZI of “conventional” low-Q, high anisotropy PN and typical 1-3 epoxy-filled PZT piezocomposite, both used to provide similar acoustical performance, with low acoustical impedance, high damping and radial mode suppression.

In the present research the “no oil” condition is provided with polarization in “air” (for example, under high air pressure, etc.), or “burning” the oil inside after traditional polarization in oil. The basic effect for oil saturation, during polarization process or after, is considered as a capillary effect. In its classical description, if the diameter of a tube (or a gap) is sufficiently small, then the combination of surface tension (which is caused by cohesion within the liquid) and adhesive forces between the wetting liquid and the tube act to penetrate the liquid inside the porous structure.

B. Transition of the PN Resonances between "no oil" to "with oil" Conditions

Comparative experimental electrical impedance spectra for the planar (radial) and thickness modes, with their fundamental and third-order harmonics, in lead metaniobate disk resonator were measured and considered under two conditions: saturated with oil during polarization process ("with oil") and without oil after high temperature specimen treatment ("no oil"). Same disk resonator OD 23 x H 2.5 mm was used in the experiment, and respective data are shown in Fig. 2.

The high temperature treatment ("no oil" condition) was provided with 260^{o}C soak of the sample for several hours. The treatment temperature is significantly less the PN Curie point T_c, and is determined basically based on the oil properties, taking into account some minor influence of the porous piezoelement plate thickness.

Note that there are no lateral modes in PN as in a regularly structured PZT 1-3 piezocomposite [25]. As can be estimated from the resonances shifts, PN is getting significantly at least near 40…50% stiffer when saturated with oil, and the Q-factor is going dramatically down from near 400 to 20.

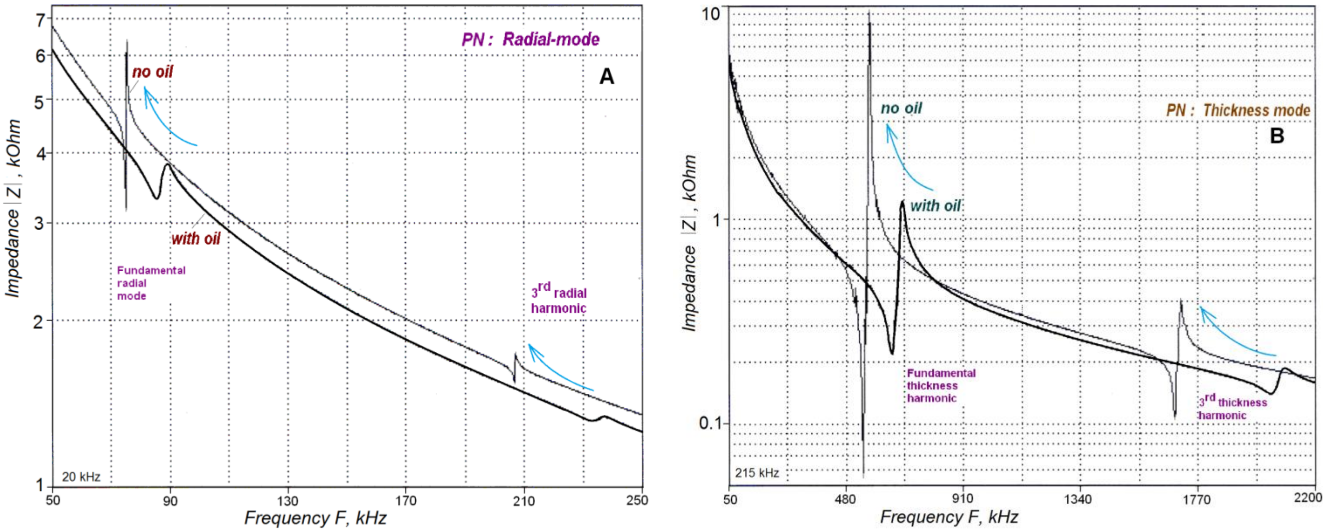


Fig. 2 A,B. Comparative experimental electrical impedance spectra for (a) planar (radial) and (b) thickness vibrational modes (fundamental and third-order harmonics) in lead metaniobate disk resonator under two transitional conditions: without and saturated with oil.

The transient resonance characteristics (fundamental modes) in the process of oil saturation were investigated in more details. Originally the polarized samples with "no oil" were prepared, and then a thin layer of oil (~0.2 mm) was put on the upper major surface of the PN disc. As seen from Fig. 3, initially there are sharp peaks in both vibrational modes with Q-factor near 300; during the first minute being in direct contact with the oil the Q-factor is dropping down significantly; and then during the next near 5 min the resonance frequency increases, with further Q-factor decreasing till fully oil saturation. The impedance analyzer was triggered in 5…30 sec time intervals, total time is near 5 min.

Similar time-characteristics behavior was noticed for the other major vibrational modes, such as rod, bar, shear types, but with some less frequency variation. Presumably the basic mechanism of the resonances shift is related to “capillary” and oil flow effects.

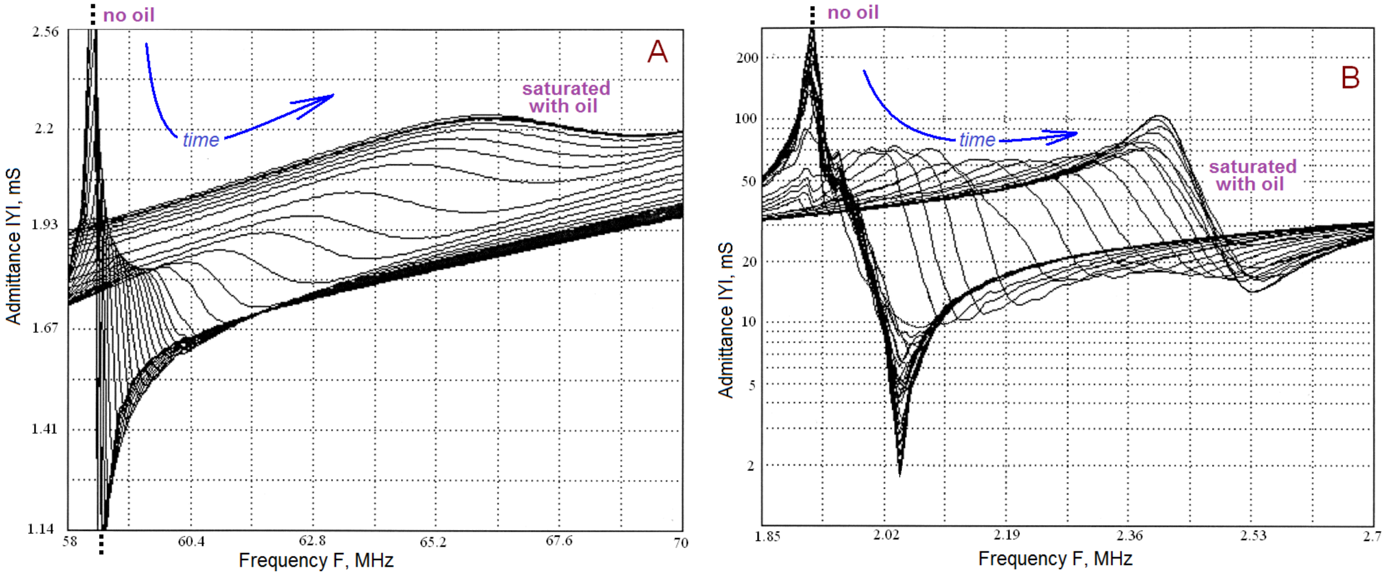


Fig. 3. Comparative transient transformation of the fundamental resonances under oil saturation for (a) planar (radial) and (b) thickness vibrational modes in PN disk resonator. The resonator OD 30 x H 0.72 mm was conventionally poled in oil, then thermo-treated to “burn” the oil (260ºC / 8 hours), and finally a layer of oil was put on one major surface during the experiment.

Notice a difference if compared to the conventional 1-3 PZT piezocomposite as a reference. When a PZT disk is cut on multiple long thin rods, and then the structure filled with epoxy, the radial resonance mode decreases significantly because of lower planar piezocomposite stiffness determined predominantly by epoxy, meanwhile the thickness resonance mode decreases a little just because of less stiffness ($1/S_{33}^E$ vs. C_{33}^E) and higher coupling coefficients CEMC (rod k_{33} vs. plate k_t) normally in piezoceramics [23].

The described oil saturation effects were FEA simulated in COMSOL with a model for a PN disk resonator describing its frequency spectra near the fundamental radial and thickness modes in the “no oil” to “with oil” PN transition, and their even harmonics. Two oil transient configurations: a) excessive amount of oil was put just on one major PN plate surface (Fig. 4 A,B, for the radial and thickness vibrational modes); b) some excessive amount of oil was put just along a part of the disk perimeter (Fig. 5). The respective material electro-mechanical constants used in the simulation for both PN piezoceramic conditions are presented in the Table I. A good matching of the simulation, compared to the experimental data Fig. 3, takes place.

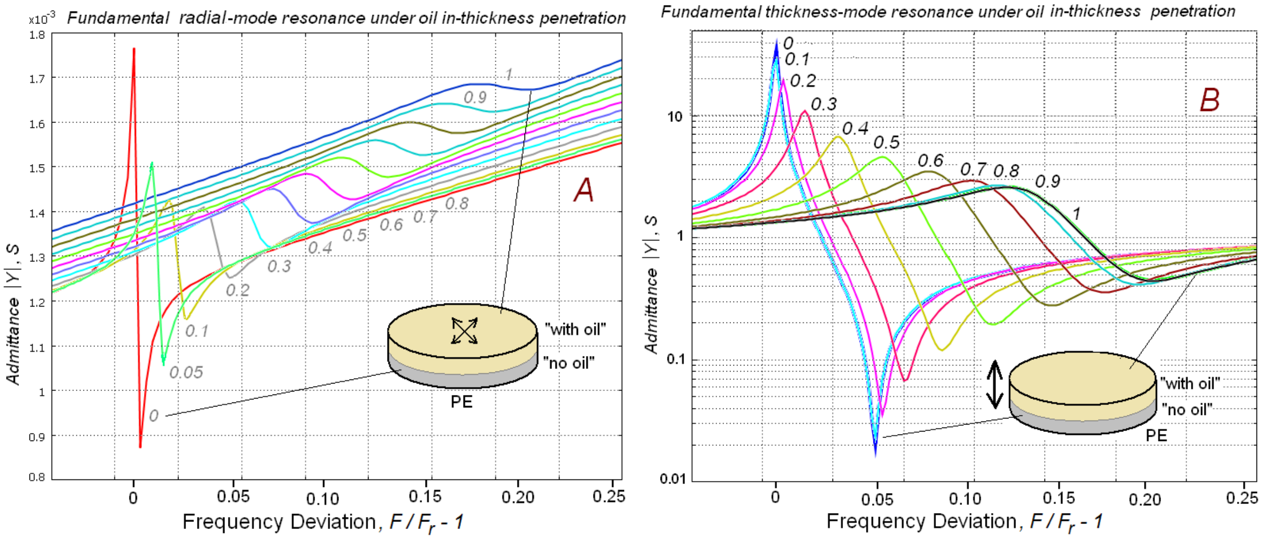


Fig. 4. Simulation of electrical admittance transformation for the (a) fundamental radial and (b) thickness modes, with partly saturated oil just inside a pre-surface layer on the major surface. Disk OD 25 with 0.5 and 5 mm thickness, respectively; Q-factor 400 and 20 for "no oil" and "with oil" conditions, respectively; the shown numbers are relative thickness of the layer saturated with oil; the arrows show the vibrational type .

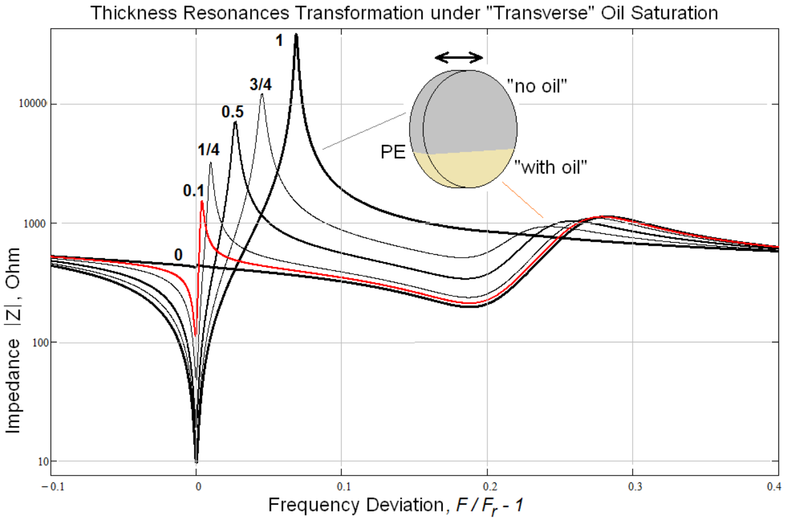


Fig. 5. Simulation results for the fundamental thickness mode, with oil partly saturated along the disk perimeter. PN disk OD 15 x H 2 mm.

C. Experimental Observation of an Even (Second) Resonance Harmonic – an Important PN Oil-Saturation Transient Characteristic

An even harmonic is not normally piezoactive, however it can be excited in the presence of structural asymmetry of any kind, such as uneven distribution of density, polarization, stiffness, etc.

A good example of the effective even-order (second) harmonic excitation is a classical Rozen-type piezotransformer [16] – a piezoelectric plate with just partly metalized major surfaces. As demonstrated in Fig. 6, all odd and even modes can be electrically exited effectively due to uncompensated charges (blue color) on the electrodes. Particularly for a PN piezoelement with thickness mode, as was mentioned the *d*₃₃-piezocoefficient is independent on oil saturation, however structurally the speed of sound varies a lot shifting the vibrational node from its symmetry point, and making the even-order resonance piezoactive.

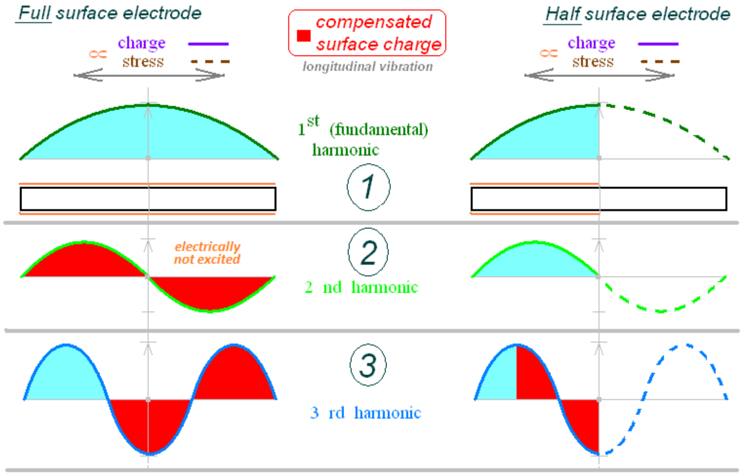


Fig. 6. Structural asymmetry with partial electrode plating in a piezoceramic rod and 2nd harmonic electrical excitation efficiency.

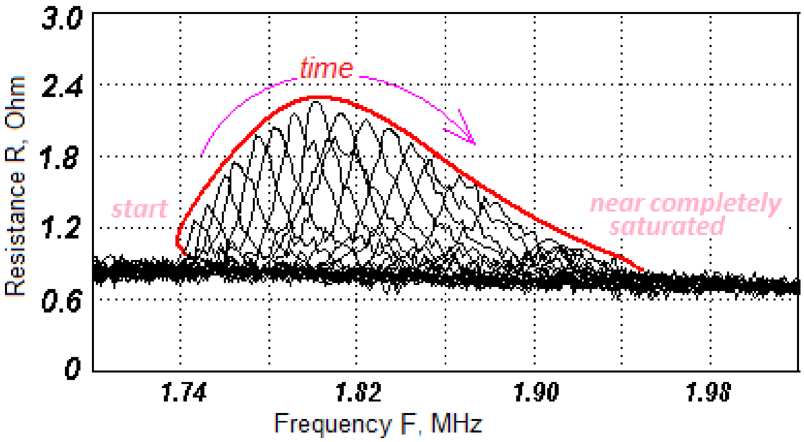


Fig. 7. Experimental data of the oil saturation effect on intensity of the second (even) thickness harmonic in PN plate piezoelement OD 30 x H 0.7 mm vs. time.

The transient behavior of the second thickness mode harmonic under oil saturation in PN is shown in Fig. 7. The piezoplate and its oil saturation conditions are similar to the ones described earlier for the fundamental modes (Fig. 3) - oil was put on a major surface of the plate initially without oil inside; the resistance *R* was recorded with triggering interval of near 0.5 min.

The growth and falling of the second harmonic intensity can be divided on the following basic stages: it starts at a lower frequency, then rise (at high oil distribution gradient), reaches a maximum (when near a half of the plate thickness is oil filled, with maximum asymmetry), then is going down relatively smoothly when the oil reaches the opposite major surface, and finally the resonance peaks completely disappears under even oil distribution along the plate thickness.

A similar experiment was conducted with alcohol saturation, as a wetting liquid (Fig. 8). Alcohol was put on a major surface of a PN plate (initially with no oil), the impedance resistance was triggered with near 0.5 min intervals. The growth of the second harmonic intensity similarly started at a lower frequency, then rises reaching near maximum (at maximum gradient / asymmetry), then is going down back to zero because of alcohol easy evaporation, finally disappearing with full alcohol evaporation. In Fig. 9 the difference in oil and alcohol penetration is schematically described.

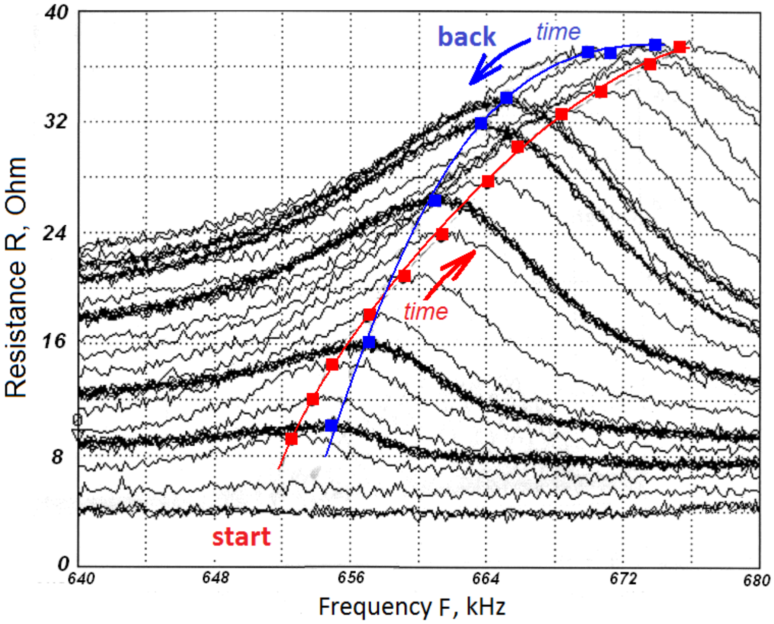


Fig. 8. Experimental data for alcohol saturation effect in time, with the second (even) harmonic of the thickness mode excitation in PN plate piezoelement OD 25 x H 2 mm.

For the pores saturation in PN, the solid(PN)-liquid wetting and capillary surface tension are the most critical. There is no a similar effect with water because of its very poor PN wetting. For different oils, particularly with higher viscosity like Molykote 200-200, the described saturation effect lasts longer.

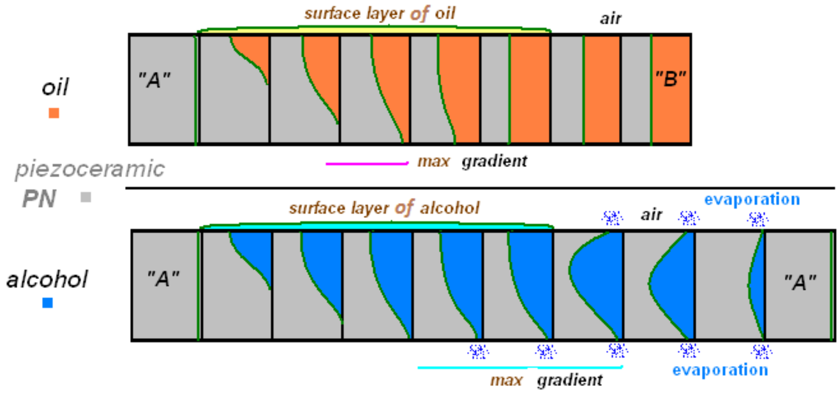


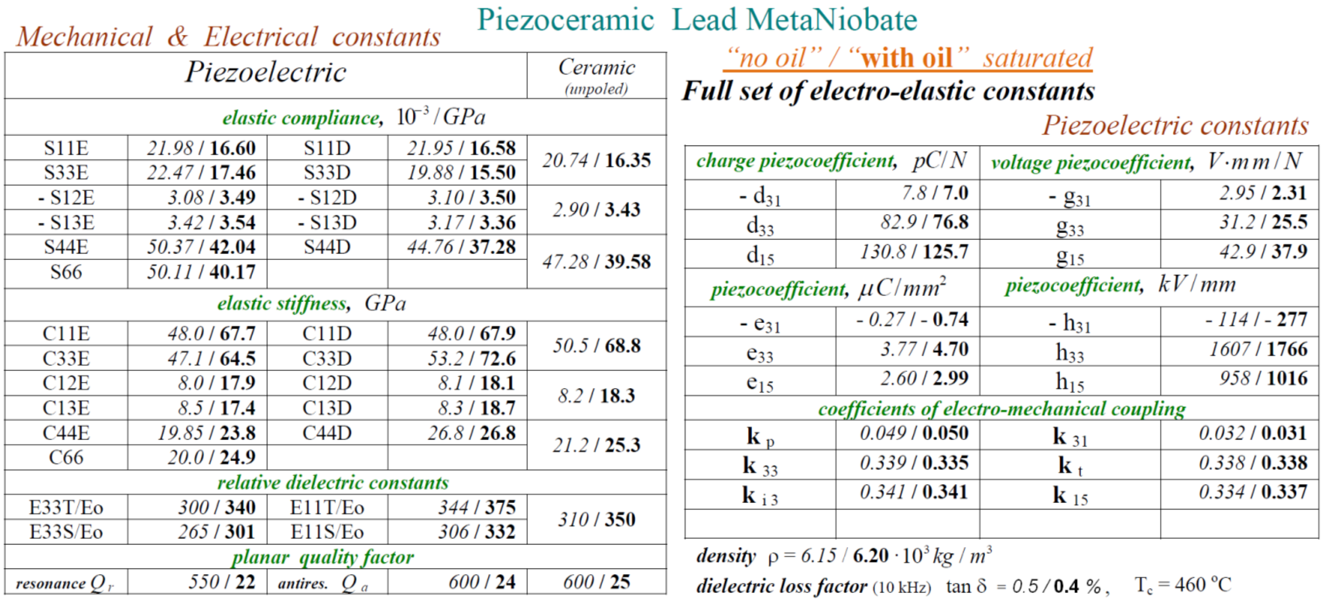
Fig. 9. Schematic for the oil and alcohol penetration inside PN plate - thickness distribution difference.

III. MATERIAL CHARACTERIZATION AND PN ELECTRO-ELASTIC CONSTANTS

A. *Electro-Elastic Constants of PN "with oil" and "no oil"*

Full set of "macro" electro-mechanical constants of PN: poled and unpoled; without oil and saturated with oil – all under normal conditions, was determined on standard samples and procedure [2], with high damping adaptation based on [16], and the results are presented in Table I. The term "unpoled" [16] is used for the samples slightly polarized with the d_{33} piezocoefficient not higher 10 *pC/N*, near ten times less the PN normal value.

TABLE I. FULL SET OF "MACRO" ELECTRO-MECHANICAL CONSTANTS OF LEAD META-NIOBATE: POLED AND UNPOLED; NO OIL AND SATURATED WITH OIL – ALL UNDER NORMAL CONDITIONS.



Mechanical & Electrical constants

Piezoceramic Lead MetaNiobate

Piezoelectric				*Ceramic* *(unpoled)*
elastic compliance, $10^{-3}/GPa$				
S11E	*21.98* / **16.60**	S11D	*21.95* / **16.58**	*20.74* / **16.35**
S33E	*22.47* / **17.46**	S33D	*19.88* / **15.50**	
- S12E	*3.08* / **3.49**	- S12D	*3.10* / **3.50**	*2.90* / **3.43**
- S13E	*3.42* / **3.54**	- S13D	*3.17* / **3.36**	
S44E	*50.37* / **42.04**	S44D	*44.76* / **37.28**	*47.28* / **39.58**
S66	*50.11* / **40.17**			
elastic stiffness, *GPa*				
C11E	*48.0* / **67.7**	C11D	*48.0* / **67.9**	*50.5* / **68.8**
C33E	*47.1* / **64.5**	C33D	*53.2* / **72.6**	
C12E	*8.0* / **17.9**	C12D	*8.1* / **18.1**	*8.2* / **18.3**
C13E	*8.5* / **17.4**	C13D	*8.3* / **18.7**	
C44E	*19.85* / **23.8**	C44D	*26.8* / **26.8**	*21.2* / **25.3**
C66	*20.0* / **24.9**			
relative dielectric constants				
E33T/Eo	*300* / **340**	E11T/Eo	*344* / **375**	*310* / **350**
E33S/Eo	*265* / **301**	E11S/Eo	*306* / **332**	
planar quality factor				
resonance Q_r	*550* / **22**	***antires.*** Q_a	*600* / **24**	*600* / **25**

"no oil" / **"with oil"** *saturated*

Full set of electro-elastic constants

Piezoelectric constants

charge piezocoefficient, pC/N		***voltage piezocoefficient***, $V \cdot mm/N$	
$-d_{31}$	*7.8* / **7.0**	$-g_{31}$	*2.95* / **2.31**
d_{33}	*82.9* / **76.8**	g_{33}	*31.2* / **25.5**
d_{15}	*130.8* / **125.7**	g_{15}	*42.9* / **37.9**
piezocoefficient, $\mu C/mm^2$		***piezocoefficient***, kV/mm	
$-e_{31}$	*- 0.27* / **- 0.74**	$-h_{31}$	*- 114* / **- 277**
e_{33}	*3.77* / **4.70**	h_{33}	*1607* / **1766**
e_{15}	*2.60* / **2.99**	h_{15}	*958* / **1016**
coefficients of electro-mechanical coupling			
$\mathbf{k}_p$	*0.049* / **0.050**	$\mathbf{k}_{31}$	*0.032* / **0.031**
$\mathbf{k}_{33}$	*0.339* / **0.335**	$\mathbf{k}_t$	*0.338* / **0.338**
$\mathbf{k}_{i3}$	*0.341* / **0.341**	$\mathbf{k}_{15}$	*0.334* / **0.337**

density $\rho = 6.15 / \mathbf{6.20} \cdot 10^3\,kg/m^3$

dielectric loss factor (10 kHz) $\tan\delta = 0.5 / \mathbf{0.4}\,\%$, $T_c = 460\,°C$

A specific feature of high-anisotropy ($|d_{33}/d_{31}| \gtrsim 8$) piezoelectrics, particularly PN, is that all e_{ij} and h_{ij} piezocoefficients are positive. As a database, the full complex of material parameters (Table

I) can be used, for example, in FEA (COMSOL, etc.) simulation of transducer acoustical behavior.

For an isotropic non-polarized ceramic state, the Young modulus and Poisson coefficient for "no oil" / "**with oil**" are: *48.2* / **61.2** GPa and *0.14* / **0.21**, respectively. Note that the relatively low effective Poisson coefficient, especially under "no oil" condition, is a consequence of high porosity [26].

Based on the "macro" PN parameters (Table I), a simplified general estimation can be proposed for the "micro" level constants of PN (as if it's without internal porous), which can be used for PN "internal" performance and analysis. The effective stiffness in the rod (k_{33} and k_{31} modes) to plate (k_t mode) transition is determined just by the internal transverse coupling, ultimately proportional to the Poisson coefficient:

A. For the "no oil" condition in PN with porosity as a rod-structure, both thickness and planar frequencies are approximately determined by the macro $1/S_{33(11)}$ (Young modulus) stiffness of the pure "micro" ceramic.
B. For the saturated with oil PN, as a more homogeneous structure, both thickness and planar frequencies are basically determined by the $C_{33(11)}$ - stiffness of the pure "micro" ceramic, with a clamped transverse vibrational coupling.

Then, the Poison coefficient of pure "micro" ceramic can be roughly estimated from the ratio of the effective (Table I) elastic constants $C_{33\text{(with oil)}}$ / $(1/S_{33})_{\text{(no-oil)}}$ ~ 1.5, which corresponds to the pure "micro" ceramic Poisson coefficient 0.33 typical for the conventional piezoceramics.

So, three levels of necessary PN characterization were considered: "micro" for "inside" performance characterization (as if no porosity), and "macro" for "outside" performance characterization; the latter with "no oil" and "with oil" saturated (Table I).

B. Range of PN Parameters Variation under Oil Saturation

The "no oil" condition is a basic reference point for the PN oil saturation effect analysis under different conditions and applications.

In general, as seen from the data Table II, the quality factor (near equally for all vibrational resonance modes), thickness mode frequency constant $N_{r\text{-}th}$ and resonances temperature stability demonstrate the highest relative variations. Particularly, as a consequence, the (dynamic) Young modulus Y increase is near 40…50%, so that the PN is getting significantly stiffer with saturated oil. Notice that the density increase is less than 1%. As described in the literature [18], PN piezoelements saturated with epoxy, then cured (it’s closer to a monolithic structure), showed the radial mode frequency constant increase up to 34% , with Q-factor near 120.

The frequency constants variations of different vibrational modes in PN under oil saturation are similar in character but different in value – the variation of the thickness resonance $N_{r\text{-}th}$ is persistently higher than for the radial $N_{r\text{-}rad}$, and at the same time the lowest variation is shown by the longitudinal (rod) vibrational mode $N_{r\text{-}rod}$. Meanwhile, all vibrational modes show extremely large deviation in the resonance and antiresonance frequencies under temperature variation in the range from 20^{o}C up to 200^{o}C, when oil is getting significantly thinner, from typical -0.5% in PN without oil to near -7% when saturated with oil (Figure 13).

TABLE II. BASIC PARAMETERS VARIATION IN PN UNDER “NO OIL” - “WITH OIL” TRANSITION.

Parameter	“no oil”, after 280’C/4days treat	“with oil”, initial	variation as “with oil” / “no oil”
Freq.constant $N_{r\text{-}rad}$, m/s	1690	2010	19% raise
Freq.constant $N_{r\text{-}th}$, m/s	1330	1680	**26%** raise
Q-factor $Q_{r\text{-}rad/th}$	400…600	**22**	**20** times drop
Piezocoeff. d_{33}, C/N	80	80	no changes
Diel.permit. $E_{33}{}^T/E_o$	300	344	15% raise
Frequency-temperature reversible variation, 20…200^{o}C	0.5 %	**7 %**	**15** times raise
Q-factor temperature variation, 20…200^{o}C	stable	stable	similar stable

Same as noticed in the literature, piezoactivity such as piezocoefficients (d_{33}) and electro-mechanical coupling coefficients (k_t, k_p, etc.) is stable. Staring after near 200^{o}C, there is an extremely high raise of “static” dielectric losses, mostly at low frequencies. As a consequence, the effective electric capacitance is going up as well. Despite the high Curie temperature point over 450^{o}C, particularly because of the latter effect, a transitional limit temperature 200…300^{o}C can be considered as problematic for some PN applications.

C. Quality Factor Frequency Dependence in Conventional PN

Q-factor of the PN saturated with oil is extremely low and relatively stable with temperature. Moreover, the Q-factor frequency dependence (including at different vibrational modes) reflects a significance of the oil dynamic (flow) performance in the piezoelectric structure. Based on the experimental data shown in Fig. 10, at relatively high ultrasonic frequencies the Q-factor increases with frequency. At low infrasonic frequencies, the Q-factor (together with the elastic compliance) increases as well, and as reported in [23] significantly, when frequency decreases. The latter occurs at the frequencies much lower the flow relaxation characteristic frequency ([27], different from the acoustical attenuation characteristic frequency) inversely proportional to the fluid viscosity. Indeed under that condition, the fluid can freely relax in the body for the time of a full excitation cycle, creating minimal stresses and additional energy losses – so that the behavior of the total PN structure is closer to its "no oil" properties, with higher Q-factor and lower elastic stiffness. Particularly for this reason, uneven oil distribution supposedly does not create extra stress gradient inside the PN sample body being under quasi-static mechanical loading.

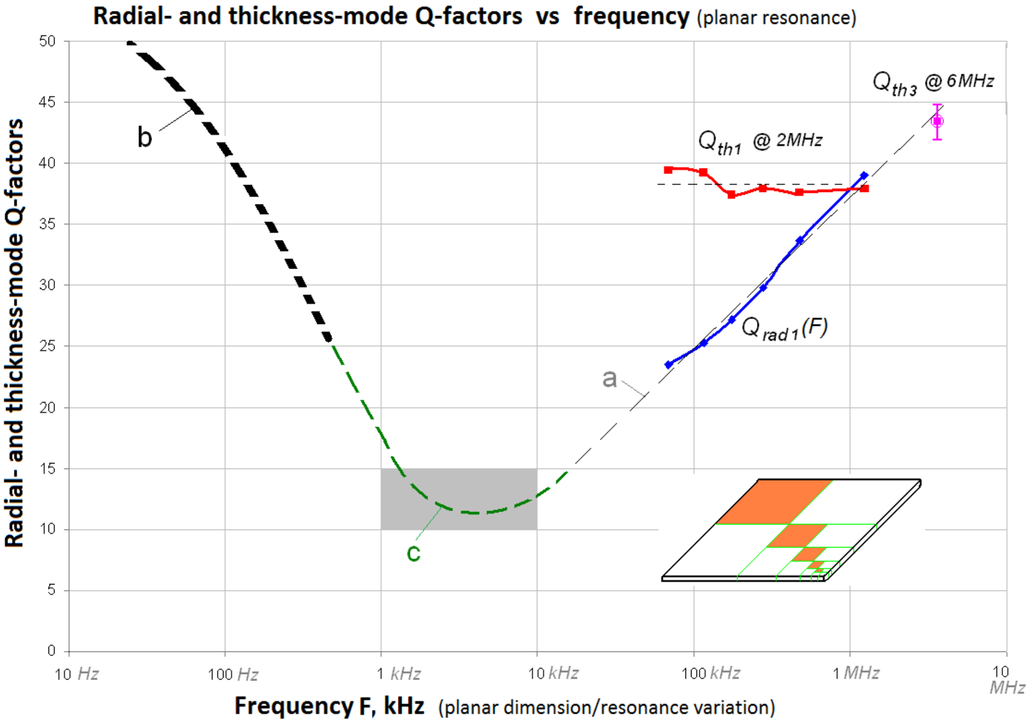


Fig. 10. Quality factor vs. frequency for PN piezoceramic saturated with oil, measured on square piezoelements with descending planar dimensions. Extra curves: *a* – linear approximation of the radial Q-factor experimental data; *b* – schematic data from the literature [23]; *c* – interpolation for curves *a* and *b*.

In the experiment (Fig. 10), initially a relatively large square conventional PN resonator, and then its smaller consecutive planar cuts as well, were taken to measure the thickness (including its higher harmonic) and planar Q-factors, with varying planar resonance frequencies inversely proportional to the planar dimensions, and constant thickness resonances.

As known, silicon oil exhibits viscoelastic behavior such that its moduli are significantly frequency and temperature dependent. Oil that behaves like a Newtonian liquid at low frequencies can act as a nearly elastic solid at high frequencies. In between those extreme cases, the wave propagation in heavy oil is dispersive and exhibits strong attenuation. Viscoelastic behavior of the oil also violates

Gassmann theory. Indeed, the classical theory is based on Pascal's Law, which states that, in the absence of body forces, fluid pressure is the same throughout the pore space. This law is not applicable to viscoelastic media. Biot's theory [28] is not applicable to heavy oils either because it ignores the viscoelastic phenomenon by neglecting the fluid shear stress in the microscopic pore-scale constitutive equations.

VI. TEMPERATURE DEPENDENCES OF BASIC ELECTRO-MECHANICAL PARAMETERS OF PN WITHOUT OIL VS. CONVENTIONAL SATURATED WITH OIL

Maximum PN operation temperature is directly related to the intrinsic PN properties [29,30], and their oil-saturated behavior is critically sensitive to the time exposure. One of the characteristic PN features is its rapid DC conductivity increase starting at 200…250^{o}C. Fig. 11 shows the effective dielectric permittivity and losses in PN without oil. Low-frequency losses are higher, and sky-rocketing after reaching 200^{o}C, getting abnormally high at 250^{o}C. The effective capacitance increases too, that basically is a direct influence of the dielectric losses increase. Note that there is no noticeable frequency dependence of dielectric permittivity and losses in PN saturated with oil up to 200^{o}C, and the dielectric loss factor is essentially small, less than 1% particularly at 1kHz.

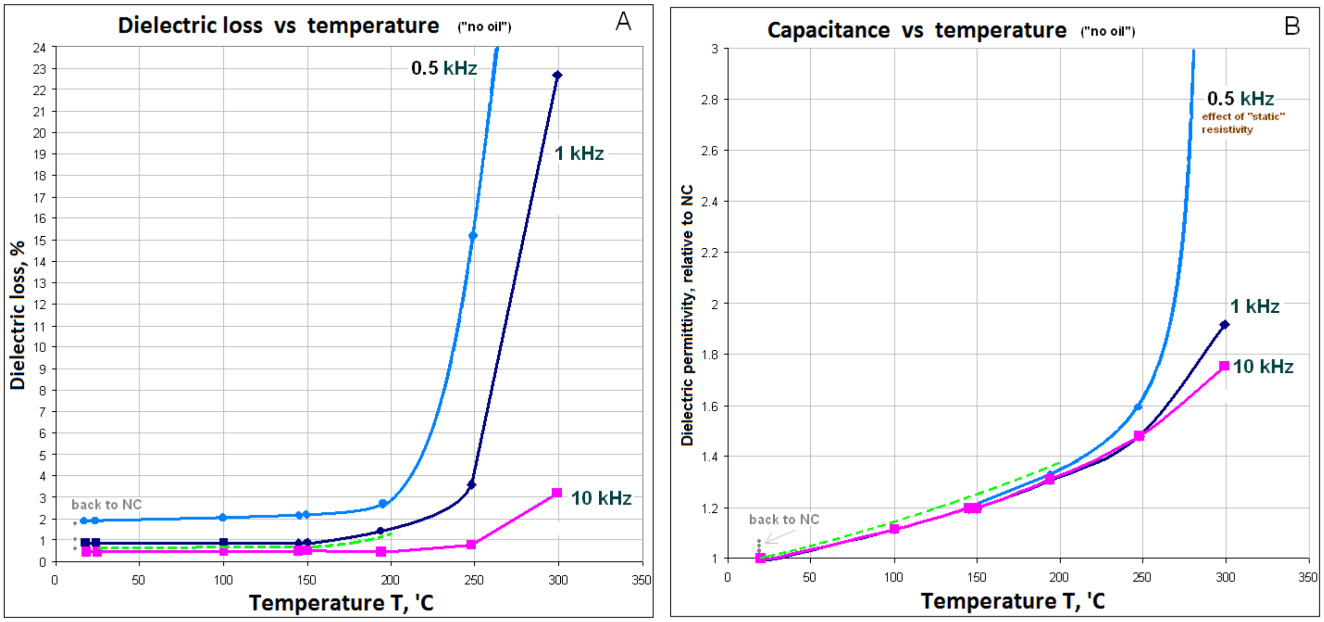


Fig. 11. Temperature dependence of the (a) losses and (b) effective dielectric permittivity at several low frequencies up to 10 kHz for PN without oil inside. For comparison, similar data for the conventional PN (saturated with oil) are presented as a green dash line.

Quality factor *Q* temperature dependence in PN saturated with oil is critically sensitive to the high temperature exposure. Particularly at 280^{o}C for 4 days soak, initial silicon oil in PN was "burned" under that treatment, with no d_{33} changes noticed. The Q-factor was permanently monitored inside the chamber, and finally the sample was slowly cooled to normal conditions. The Q-factor was measured with *X&B* frequency components of the impedance *Z*(*Y*). As seen from the data Fig. 12, after high temperature soak the PN Q-factor is significantly higher more than an order of magnitude (150…400, depending on sample fixture conditions in the chamber), and stable on temperature.

Variation of resonance and antiresonance frequencies for both PN conditions (without and saturated with oil) is presented in Fig. 13, from normal conditions up to 300/200°C, respectively. Both radial and thickness frequencies are as much as near 10 times more stable with temperature for the pure PN without oil compared to the oil-saturated condition. Note that the relative resonance intervals as a measure of piezoactivity remains stable in the both cases.

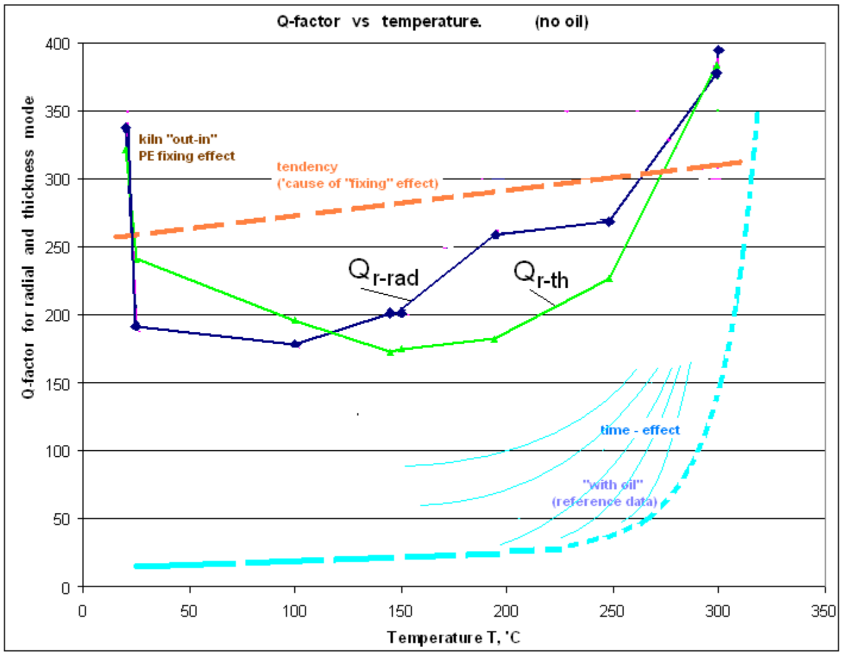


Fig. 12. Quality factor temperature dependence in PN with “burned” oil (4 days soaking at 280°C) inside. Under Q-factor permanent monitoring, the sample then was slowly cooled to normal conditions. Sample OD 20 x H 0.72 mm; radial mode 58.5 kHz and thickness mode 2.5 MHz.

For the oil-saturated PN ceramic under elevated temperatures higher 200°C, the test result depends on the time-temperature test conditions, so that being some longer under high temperature results in the frequencies dropping down significantly because of the oil phase transformation; meanwhile the Q-factor increases, but is not so sensitive to relatively short-runs (Fig. 14).

When a solid PN skeleton is saturated with heavy oil, its behavior also becomes viscoelastic, particularly with a dramatic decrease of compressional velocities in an oil-saturated body with increasing temperature. The main factor responsible for that velocity decrease is variation in the fluid properties with temperature, similarly for transverse and longitudinal vibrational modes – oil is getting much thinner at higher temperatures, with significantly lower viscosity. Such an effect does not take place in the conventional 3-1 PZT piezocomposite, with significantly more stable thickness modes.

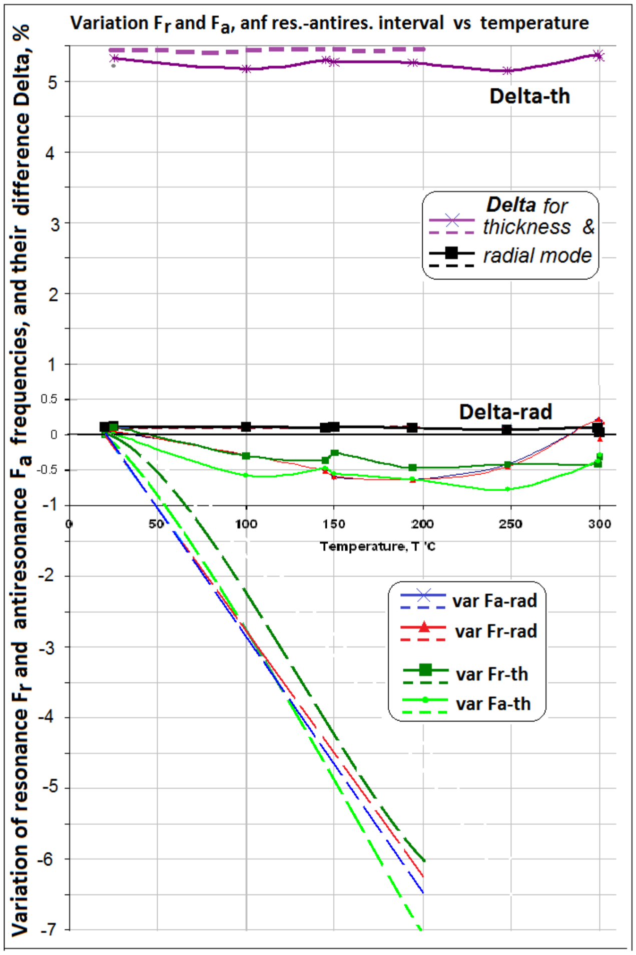


Fig. 13. Comparative temperature variations of the basic resonance parameters (resonance/antiresonance frequencies, and their respective differences (*Delta*)) at the thickness and radial modes in lead metaniobate with (solid lines) and without oil (dashed lines).

PN saturated with oil has a relatively low Q-factor and it is stable with temperature at least up to 200^{o}C, with conventional test soak conditions just for several hours (Fig. 14). That fact can be explained based on the dynamic viscous flow of oil in the PN porous structure. The losses of the vibrational energy are proportional to the product of the internal pressure times flow velocity. If the internal deformation, as a stored energy factor, is a direct consequence of the oil redistribution (its velocity), then it provides stable Q-factor behavior in the porous structure under oil viscosity variation with temperature.

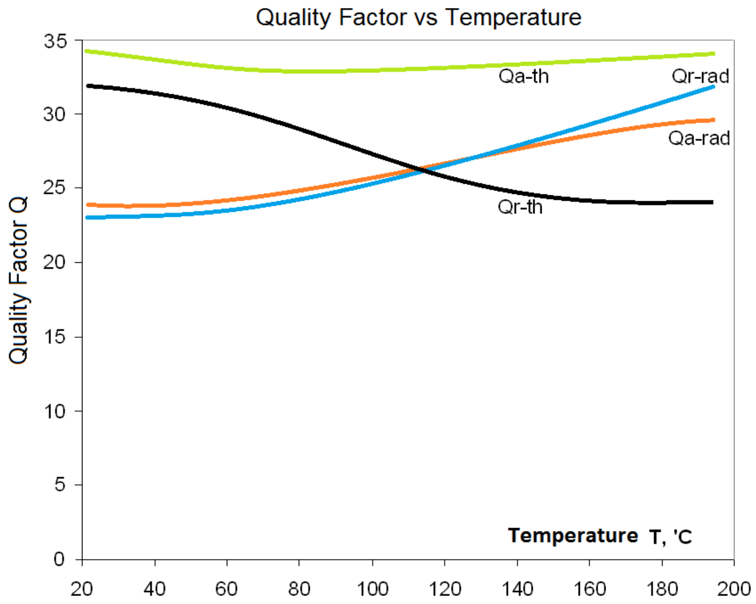


Fig. 14. Quality factors temperature variation at the thickness and radial modes in PN saturated with oil under standard performance-over-temperature test.

V. SILICON OIL THERMAL PROPERTIES UNDER HIGH TEMPERATURE SOAK TREATMENT (260^{O}C) AND ITS EFFECT ON TRANSIENT CHARACTERISTICS IN PN BEING SATURATED WITH OIL

A. Silicon Oil High Temperature Behavior

Two separate portions (3…5 ml) of initially liquid silicon oil (Fig. 15A) were soaked at 260^{o}C: both the portions were put in small aluminum cups, the first portion was covered with an aluminum plate, the other one was open during all treatment. The oil conditions were periodically monitored. Chronologically, the *open* oil got very thin with high fluidity, the surface was fuming (first 4 hours); after some characteristic moment (near 6 hours) it got thicker, then got like a gel (like a jellyfish) with no flow possibility (after 8 hours); then was getting harder, like ice or glass (after 16 hours), and very fragile; finally it got solid (Fig. 15C) and "sounds" like a bell (after 1.5…2.5 days). Total shrinkage was noticed near 2…3 times in volume. The other *closed* oil was of very high fluidity liquid (8 hours), then like a gel in 16 hours, and finally like a gum in 2.5 days (Fig. 15B). An reverse process was investigated: several like-gum and like-glass pieces of the previously tested oil were put into regular oil – after a day of treatment under room temperature and 130^{o}C, nothing essential happened to the pieces.

Based on the data, in respect to the conventional PN piezoceramic (saturated with oil), it was concluded that the expected oil characteristic time of the fluid-solid transformation (polymerization) is several days, and the effect can be critical particularly to the piezoelement thickness (for standard disks).

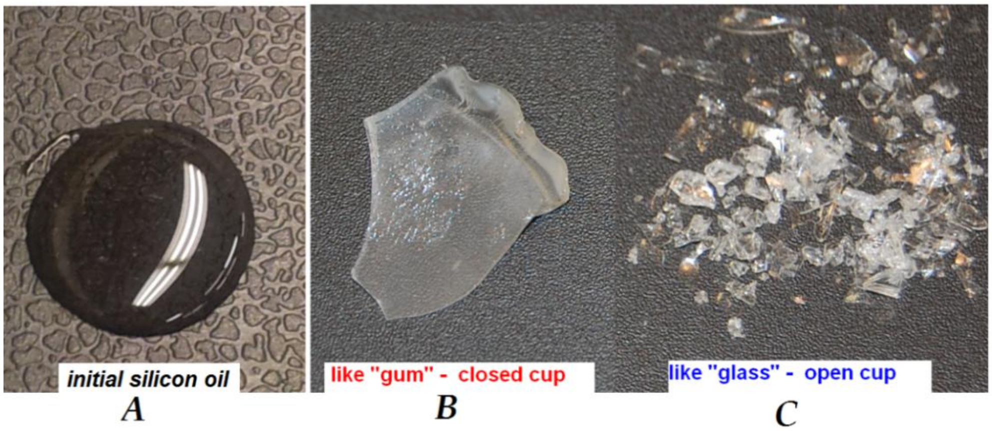


Fig. 15. Oil consistency transformation in an experiment: from (a) initially liquid then (b) to like a gum and (c) finally glass conditions takes near 2.5 days in high-temperature soaking.

The silicon oil transformation is also known as baked-on siliconization and widely used in practice in other industrial applications. Particularly, it involves the application of silicone oil as an emulsion which is then baked on to the glass surface in a kiln at a specific high temperature and for a specific length of time. The bonds are so strong that the solid silicone can not be removed with solvent. A permanent extremely thin hydrophobic layer is then created [31,32] – the average silicon molecule weight increases as a result of polymerization and vaporization of short chain polymers. Baked-on siliconization reduces the measurable quantity of initially free silicone oil to approximately 10 % of the normal value.

B. Reversibility and Transient PN Characteristics under Oil Saturation

The data for irreversible variation of the basic properties in PN treated under 260^{o}C is presented in Fig. 16. A disk 20x1 mm sample was soaked at that temperature for a specified time, then periodically taken out from a temperature chamber and characterized under normal conditions during near 30 minutes each point. In the chamber, the sample was wrapped in aluminum foil to provide a short-circuited electrical condition and isolate the sample to minimized evaporation of the light oil components. The total treatment time took 10 days.

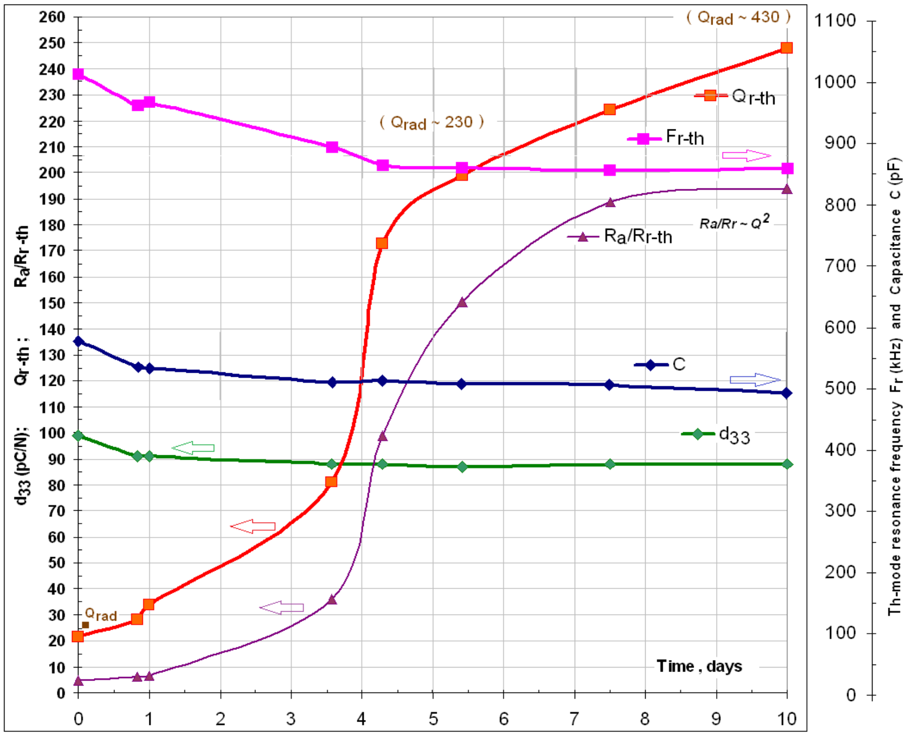


Fig. 16. Irreversible PN parameters variation under 260ºC soak, all measured at normal conditions: d_{33} piezocoefficient, capacitance *C*, thickness mode resonance frequency $F_{r\text{-}th}$, quality factor $Q_{r\text{-}th}$ and resonance intensity $R_a/R_{r\text{-}th}$. Disk OD 20 x H 1 mm.

In general, the variation of the radial resonance frequency constant was found from 1933 → 1757 Hz·m (-10%); Poisson coefficient 0.20 → 0.14; relative dielectric constant 354 → 303 (-16%); thickness resonance frequency constant from 1615 → 1378 Hz·m (-17%). The percentage is given as a parameter ratio of the initial (with oil) to final (without oil) conditions.

Quasi-static piezocoefficient d_{33} is relatively stable, with some decrease near 10% after the first-day treatment. The Q-factor increases near linearly from 20 to 80 during the first 3.5 days of treatment; then jumped significantly up to 180 after 4th treatment day, and then increased near linearly up to 250 to the end of the final 10th day of the experiment. Thickness resonance frequency and dielectric constants decreased mostly during first 4 treatment days reaching saturation.

Two types of experiments were performed at the 260ºC temperature point: a sample was inside the temperature chamber for a definite period of time, then removed from the chamber, its parameters were measured under normal conditions, and the sample was put into the chamber for further treatment (Fig. 16). In the second experiment a sample was permanently inside the temperature chamber and its parameters variations were monitored inside the chamber (Fig. 17). The samples were permanently short-circuited under temperature variations with a 1 MOhm resistor to avoid pyro-effect, the samples were open in air.

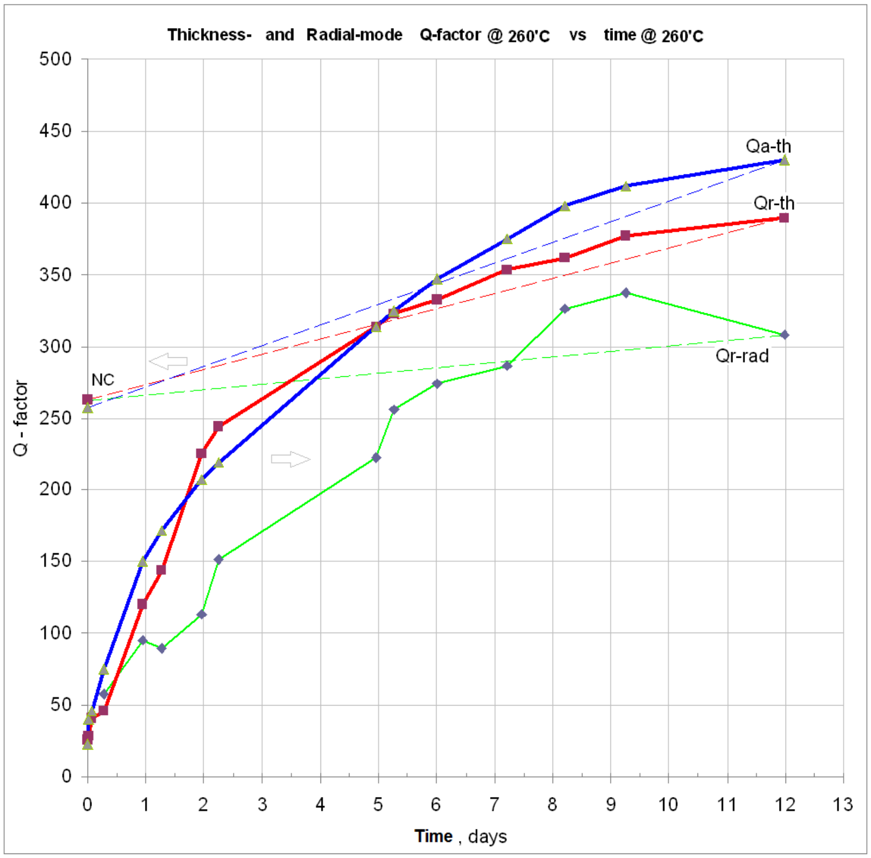


Fig. 17. PN Q-factor variation under high 260ºC temperature soak (days). The measurements are made at the current treatment temperature; the sample was open (not wrapped) and permanently connected to the circuitry.

In the latter case, the time variation of the Q-factor is near exponential, with a characteristic time near several days. Q-factor increases from 20 to 100…150 already after the first day of the thermal treatment.

So, the properties of PN porous piezoceramic saturated with oil (liquid filler), determined under both elevated temperatures and normal conditions, depend on the pre-history of the samples, and are mostly related to the oil transformation under high temperature soak. Based on that, the basic factors influencing the PN time-performance are: a) temperature and time factors, for both oil evaporation/penetration effects; b) sample thickness (surface-to-volume ratio), as a critical parameter for oil evaporation/penetration effect depending on: c) open / closed volume around the sample (evaporation conditions).

Particularly, the effect of sample thickness was investigated in more details in this respect. Comparative data with different thickness' of PN disk samples are presented in Fig. 18. Three characteristic disk samples were tested, with same diameter but different thickness' in a row 2, 3, and 5 mm. The samples were conventionally prepared; the initial Q-factor was near 25. In the experiment, the samples were short-circuited, put into temperature chamber, soaked for 4 hours (b, Fig. 18) and 28 hours (c, Fig. 18) at 260°C, and finally put into hot (80°C) oil for 30 min (d, Fig. 18). The samples were measured after each stage during near 30 min under room temperature.

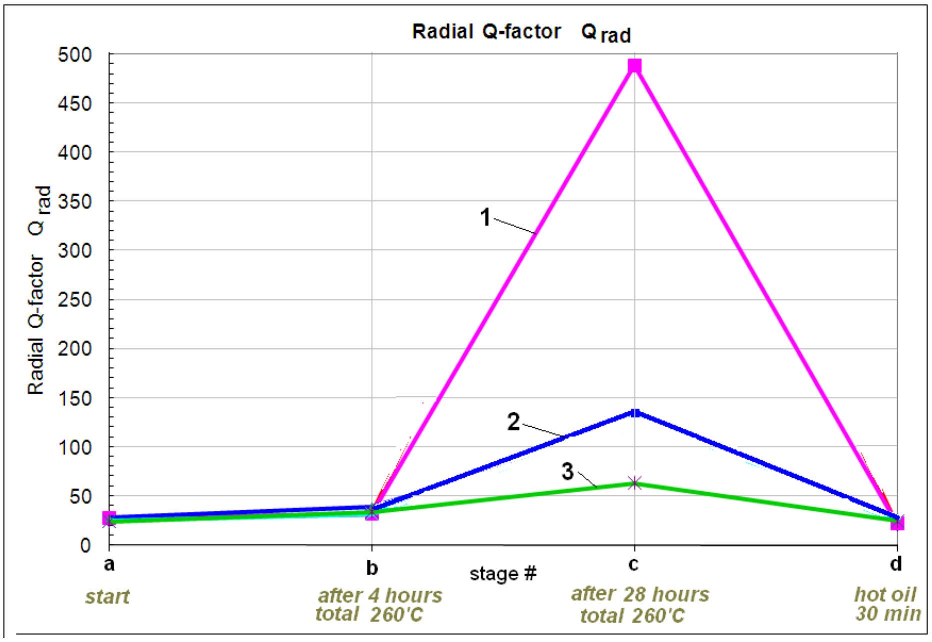


Fig. 18. PN Q-factor (radial mode) reversibility at NC under temperature treatment and final oil saturation for different piezoelement thickness': line 1 – 2 mm, 2 – 3 mm, 3 – 5 mm, of the same diameters.

After the first 4 hour treatment for all samples, Q-factor was relatively stable. Then for longer times, a strong effect of sample thickness takes place: Q-factor of the thinnest sample was increased up to 500, while in the thickest sample the Q-factor was just 70 – all after an additional day of thermal treatment. Soaking in oil returned back their low Q-factor (near 25) in all samples. Note same effect took place for the resonance frequencies and capacitance of the treated samples.

The described effect is reversible – saturated with oil again after high temperature treatment, the PN returns back to its low Q-factor condition – it was checked at least 4 times in a row.

VI. DISCUSSION THE EFFECTS RELATED TO PN OIL SATURATION

The Q-factor frequency dependence of conventional PN piezoceramic in most practical 100 kHz – 6 MHz frequency range was measured on square elements with consecutive descending planar dimensions, as shown in Fig. 10. The radial mode Q-factor at high frequencies increases approximately as $Q \sim f^{1/5.5}$, close to typical PZT, while the thickness mode Q-factor with unchanged thickness, as a control parameter for all planar dimensions, was relatively stable. Adding the data known in the literature for the conventional PN [2], minimal mechanical dissipation Q ~ 15 is expected at the frequencies 1…10 kHz, with further rapid increase at lower infrasonic frequencies.

Based on a general analysis, the Young modulus vs. frequency is expected to exhibit a similar frequency dependence as the Q-factor at low infrasonic frequencies, with transition from high stiffness at ultrasonic frequencies to lower stiffness (Table I) at least at static, because at quasi-static there is no elastic/viscosity oil effect in the porous structure with open interconnected pores.

To explain the extremely low Q-factor in PN, the following acoustical approach was used for Q-factor estimation. As known in general, in a process of transducer immersing in a medium, the contacting transducer's front emitting face to some other surrounding material provides an acoustical matching between the transducer and its acoustical load. In that case [16], the total Q-factor of a mechanically (acoustically) loaded piezoelement is determined as follows:

$$\frac{1}{Q} = \frac{1}{Q_m} + \frac{4}{\pi} \cdot \frac{Z_L}{Z_0} \cong \frac{4}{\pi} \cdot \frac{Z_L}{Z_0} \quad . \tag{1}$$

The difference of a porous PN with the classical model is that the acoustical loading liquid is located inside the vibrating porous PN element, and can be considered like an "internal" load in liquid. There are two critical aspects of such a consideration – energy transmission, which is described by (1), and dissipation of that energy in the load. The latter is basically related to the viscose liquid flow under piezocrystal alternative strain, and some portion of the energy possibly returns back to the PN crystal frame (main body). Particularly for the simplified model, it's considered as fully dissipated.

For a relatively high intrinsic quality factor Q_m near 500 of PN, the total energy dissipation in piezoelement is determined by the relatively strong oil acoustical matching, so that the effective Q-factor of PN saturated with oil is estimated as $Q \cong \pi Z_0 / 4 Z_L = 12$, for $Q_m = 500$, piezoceramic main body acoustical impedance $Z_0 \sim 20$ MRayls and loading liquid (oil) impedance $Z_L \sim 1.3$ MRayls. Note a good agreement of the estimation with the actual conventional PN material dissipation parameter.

The dielectric permittivity variation under oil-saturation effect requires a separate consideration, for a relatively large effective capacitance increase reaching near 15% when PN filled with oil. For a comparison with a longitudinal 3-1 composite structure, with empty relative volume less than 1:10 and the filler dielectric permittivity near 100 times less than that in piezoceramics, the dielectric permittivity increase can be well below 1%.

A simplified elementary 2-D model was used for estimation of the dielectric permittivity variation in the porous PN structure, and then its internal effective gap was determined (Fig. 19). In the model, both longitudinal (in respect to the applied voltage) and transverse internal structural gaps between grains in PN were considered.

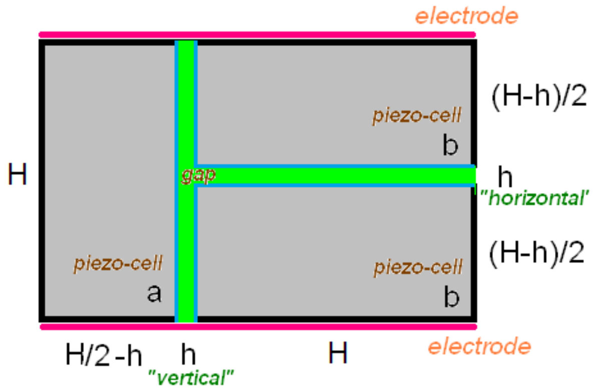


Fig. 19. A simplified 2-D model with major longitudinal and transverse gaps in elementary structural cells for the porosity effect estimation: h – gap (between grains); H – characteristic grain dimension with a 2:1 aspect ratio for certainty.

The effective dielectric permittivity of the total cell is determined through the capacitance of the model of Fig. 19 as:

$$\varepsilon_{eff} \cong \varepsilon_{piezo}\,\frac{1}{3}\left[1+\frac{2}{1+\frac{h}{H}\left(\frac{\varepsilon_{piezo}}{\varepsilon_{gap}}-1\right)}\right] \tag{2}$$

As seen from (2), maximum gap influence on the total cell capacitance is formed by the horizontal gap, and a relatively large increase near 20%, if compared the air vs. oil fillers in the gap, is provided by a relatively narrow gap with a thickness near $h/H \approx 10^{-2}$. Then for a characteristic PN grain dimension ~30 μ, an estimated capillary gap has a micron, or even sub-micron, thickness.

Based on the practical data for the oil saturation effect on the PN stiffness variation reaching near 40…50%, an attempt was made to estimate the basic intrinsic elastic parameters of the PN itself (in the skeleton). This data can be useful particularly for simulation of the PN porous structure dynamic interaction with saturating materials. Note that the effective parameters in Table I just correspond to the porous PN with no filler, and filled with oil.

First of all, the frequency constants variations under oil saturation for all vibrational modes are similar but essentially different in value – particularly the variation of the thickness mode frequency constant $N_{r\text{-}th}$ is persistently higher than in the case of the planar (radial) frequency constant $N_{r\text{-}rad}$ (Table II). If the oil is considered as incompressible, the reference limit conditions for the ratio of plate-to-rod resonances in-poling direction $m_{33} = F_{th} / F_{rod} = \sqrt{\bar{C}_{33}^{E}\bar{S}_{33}^{E}}$, and perpendicular to the poling direction $m_{11} = F_{th} / F_{bar} = \sqrt{\bar{C}_{11}^{E}\bar{S}_{11}^{E}}$, where $\bar{C}_{ij}^{E}$ and $\bar{S}_{ij}^{E}$ are respective intrinsic piezoceramic stiffness. Typically, the m_{33} and m_{11} values are 20…40%, depending on the ceramic type (PZT), and $m_{33} > m_{11}$. With such a simple consideration based on the "gap" approach, the basic intrinsic PN piezoceramic constants can be estimated. So, for the intrinsic PN constants, $\bar{S}_{33}^{E} \cong (S_{33}^{E})_{no-oil} \cong 22\ 10^{-3}$ 1/GPa ($\approx (1/C_{33}^{E})_{no-oil}$), accompanied with a relatively low effective Poisson coefficient; and $\bar{C}_{33}^{E} \cong (C_{33}^{E})_{with-oil} \cong 66$ GPa. Then, the ratio $\bar{C}_{33}^{E}\,\bar{S}_{33}^{E} = 1.45$, corresponding to the 0.33 intrinsic PN Poisson coefficient as an estimation.

Under temperature variation, the compressibility of oil (and supposedly its non-linear limits) is going up at higher temperatures [31], as particularly follows from the speed of sound temperature dependence for silicon oils, where the compressibility is inversely proportional to the speed of sound squared. PN piezoceramic itself (without filler, see Fig. 13) is relatively stable, so it can be concluded that in the PN saturated with oil at higher temperatures both frequencies are going down, up to the "no oil" limit (~20…25%) when the oil is getting relatively high compressible. At colder temperatures, the frequencies increase, with expected limit near ~5% .

Supposedly, the thermal expansion anomaly noted in [24] can also be related to the silicon oil inside the PN structure, with the oil characteristic low temperature "pour" point, when silicon is getting semi-solid with lost its flow possibilities.

VII. CONCLUSION

The full sets of "macro" electro-mechanical constants were determined for PN under two conditions: conventionally saturated with oil and without oil, which both particularly can be used for external acoustical performance evaluation of PN. Also, the intrinsic elastic and dielectric constants on a "micro" level were estimated based on the open-pores liquid saturation effects, useful for describing the internal electro-elastic performance of the PN piezoceramic.

The time transient resonance characteristics in the process of oil saturation were experimentally studied and described, and found a satisfactory explanation in the FEA simulation with a layered structure approach for both thickness and radial vibrational modes. As was found and demonstrated in the time domain, the intensity of the second (even) harmonic is a measure of the maximum structural inhomogeneity under oil penetration in the PN structure.

The temperature dependences of the basic resonance and dissipation PN characteristics were experimentally determined for both oil saturated and no-oil conditions. As known, the PN conductivity, and the effective piezoelement capacitance as a consequence, are critically rising starting at 200…250^{o}C temperature, with the strongest effect at low (static) frequencies. At the room temperature, the static conductivity and low-frequency dielectric losses are significantly lower in PN saturated with oil.

Inside the temperature interval up to 200^{o}C, the piezoelectric activity, expressed as the resonance frequency interval of a vibrational mode, is relatively and equally stable for both PN conditions without and saturated with oil. The resonance/antiresonance frequencies for both thickness and radial vibrational modes are relatively stable with temperature within 0.5% in PN without oil, and their variation is significantly increasing to near 7% in PN saturated with oil. The quality factors are essentially different near 500 and 25, respectively, and in both cases are relatively stable with temperature.

The quality factor frequency dependence in PN saturated with oil shows a typical for piezoceramic slow increase at the ultrasonic frequencies. Extrapolating these data to the sonic frequencies, and combining it with the data [23] for the infra-sonic and sonic frequencies, it was predicted a minimal Q-factor value near 10…15 at the frequency range 1…10 kHz. As follows from the present research, in the oil saturated PN, the effect of oil on the elastic stiffness and Q-factor diminishes at quasi-static (low infra-sonic frequencies), explaining the experimental data of [23].

As was experimentally demonstrated, at higher temperatures (particularly 260^{o}C soak), a transition from “with oil” to “no oil” in the PN properties takes place. As was shown in the separate experiments with pure oil, this effect is directly related to the phase transition in oil, from liquid to solid state, with a significant role of the oil light component evaporation. The characteristic transition time in PN for particularly Q-factor from near 25 to 500 is near several days, depending particularly on the piezoelement thickness (actually the ratio of the open surface to volume). The effect is reversible – saturated with oil again, PN returns back to its low Q-factor condition (it was performed at least 4 times in a row).

Summarizing, the unique extremely low Q-factor property of lead metaniobate is directly related to the oil saturation of its porous structure. Not oil-saturated PN itself is a far different piezomaterial, particularly much less attractive and valuable, with an order of magnitude higher Q-factor and near 25% shifted frequency constants. All that is critically important when any technological variations with PN are considered. That basic conclusion also explains a contradiction with the PZT composition family, when conventionally the lowest material Q_m-factors correspond to the lowest Curie points and highest dielectric constants.

The conventional PN behavior is mainly determined and restricted by oil, but knowing the details of such an effect can provide better reliable and predictable PN performance, particularly at high temperatures, and on the other hand determines the way to get even lower Q-factor. The investigated and discussed PN properties can be used as a basis for more detailed and broader FEA multidisciplinary simulation analysis, with expected new technical results for the unique structure-fluid interaction in the porous active piezoceramic.

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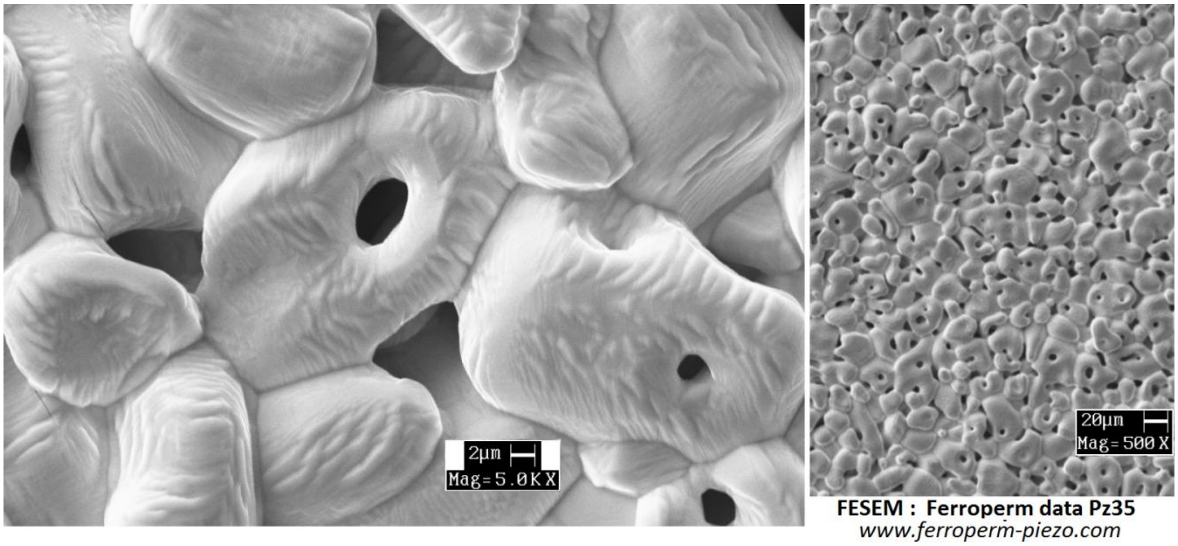
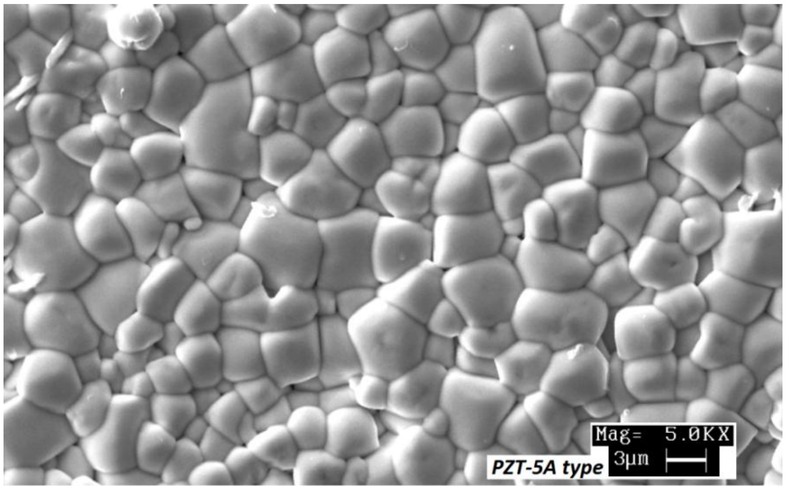
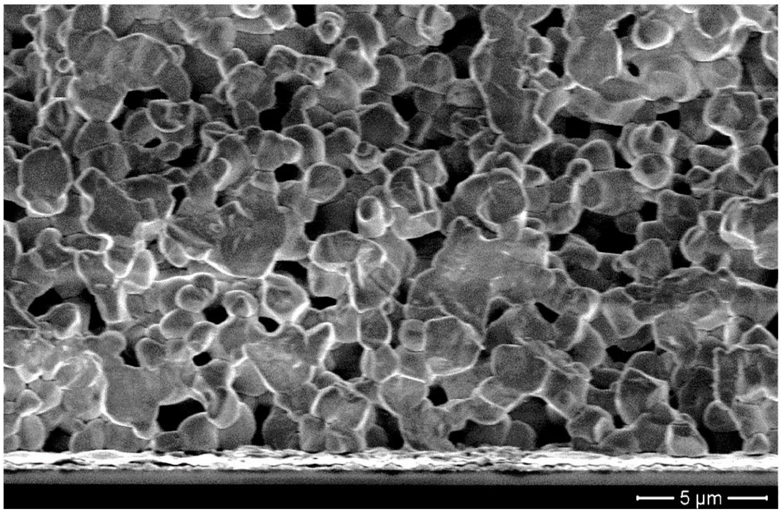
Appendix



PN (Pz35) micro-structure: the grain shape, specific of grains connection, and net of open gaps.
(by Ferroperm www.ferroperm-piezo.com, 2016)



Std PZT-5A type – for comparison (Pz27 Ferroperm).



Porous 25%; soft piezoceramic, thick film.

E.Ringgaard, F.Lautzenhiser, L.M. Bierregaard, T.Zawada and E.Molz "Development of Porous Piezoceramics for Medical and Sensor Applications," Materials, v.8, 2015, pp. 8877–8889 ; www.mdpi.com/journal/materials

same Q with oil