A Unique Fractional Derivative Operator to Simulate All Dynamic Piezoceramic Dielectric Manifestations: From Aging to Frequency-Dependent Hysteresis

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A unique fractional derivative operator to simulate all dynamic piezo ceramic dielectric manifestations: From aging to frequency dependent hysteresis.

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Abstract:

Ferroelectric materials are utilized in many applications ranging from capacitors to data storage. The hysteresis frequency dependence of ferroelectric materials has been well studied. However, the long term dynamic behaviour including aging has not been as well documented due to the long time frame required to gather experimental data, but is critical for understanding the lifespan of these materials in application. Previous work has shown that the hysteresis frequency dependence of the dielectric properties can be accurately modelled in the time domain using fractional derivative operators applied on a large frequency bandwidth. Currently, the lowest frequencies tested have been restrained to the hysteresis cycle quasi-static threshold. Below this threshold, the hysteresis shape remains unchanged. This research expands the current knowledge by validating the use of fractional derivative operators in long term aging models. The model data is experimentally validated using aged piezoelectric samples with over up to 10^7 seconds. These results confirm that the low and high dynamic dielectric material behaviours are linked and can be consequently modelled using fractional derivative operators.

Keywords : Aging, fractional derivative, frequency dependence, hysteresis, ferroelectricity, piezoelectric materials.

I. INTRODUCTION

Ferroelectricity is a material characteristic defined by a material ability to exhibit spontaneous electric polarization which can be reversed by the application of an external electric field [1]. Piezoelectricity is the ability of a material to exhibit polarization variations when subjected to a mechanical stress. Piezoelectric materials also show the converse effect, undergoing mechanical deformation under the application of an electric field [2]. These properties lead to numerous applications for piezo ceramics including sensors, actuators, and energy harvesters. However, the ferroelectric and the piezoelectric properties of a piezo ceramic are often highly non-linear, which can cause challenges in calibration and real world applications. As a result, in depth characterization of piezo-ceramics is needed. This includes characterization of piezoelectric polarization, fatigue, aging, mechanical properties, and electrical properties in accordance with frequency dependence. These properties have been investigated and characterized for use in practical applications [3]. In application, most piezoelectric devices operate under small signal conditions when compared to the initial load (high voltage) applied to pole the samples. The general operating signal is a low voltage electrical input or minor mechanical deformation. As a result of minimal loading conditions, the material's behavior stays within the linear performance range. However, when piezo materials are used as actuators, the excitation levels required are higher and can be comparable to the initial poling excitation. Precision positioning systems and mechanical vibration energy harvesters are examples of such applications [4][5]. These excitations cause reconfiguration of the domain which can lead to domain wall displacement and variation in the polarization field (P) along with non-linear responses.

By plotting these polarization field variations versus the electric field (*E*) or versus the mechanical stress (*T*), the dielectric hysteresis cycle *P(E,T)* can be observed [6]. Piezo-electrics have many advantages including their design flexibility, ability to withstand large strains, low cost, low maintenance, their ability to convert vibrations into electrical energy, pollution free energy source, and they are unaffected by external electromagnetic fields. However, there are limitations as well including the requirement of an input signal such as mechanical deformation or electrical supply. These materials are also generally temperature sensitive, and piezoelectrics are prone to cracking under high stress conditions. With the given, pros and cons in many applications, piezo-electrics are a very good solution. In all product applications, the long term performance of the materials must be considered and is generally defined as aging [7] and fatigue [8]. Aging is defined as the gradual changes in material properties with time under external equilibrium conditions. In piezo materials, aging is the usually detected by logarithmic time variations of key piezo ceramic parameters [9][10]. Fatigue is characterized by gradual changes in the material properties under loading conditions.

Aging is an inherently time dependent material based characteristic that demonstrates the material's dynamic behavior under extremely weak frequency levels.

Models based on the space charges migration have been proposed to model ferroelectric aging [11]-[13], but these models are currently unable to provide short term behavior information including properties such as dielectric hysteresis dependence (*P(E) f* ϵ [0.01-10 Hz]). In literature [14]-[17], it has been demonstrated that by using fractional derivative operators the frequency dependence of a piezo ceramic dielectric hysteresis can be correctly simulated on the full frequency bandwidth of a variety of piezo material, even including bulk ceramics and thin films.

In these sensitive setups, 3 decades can be seen for a bulk-type ceramic and 6 decades for a thin film. Creep phenomena (the piezo ceramic drift of displacement under a constant applied electric field) has also been simulated with success [18].

Under excitation frequencies larger than 10 mHz, experimental results are easier to test for and model because experimental tests can be performed in a short period of time. However, in all aging experiments environmental conditions including temperature, pressure, and humidity must be carefully maintained and monitored in order to get reliable results.

Lead based $Pb[Zr_1{-}xTix]O_3$ (*PZT*) ceramics exhibit excellent temperature stability and are highly adaptable for numerous applications because they can be easily doped with additives to adjust their electrical and material properties to meet desired specifications. Because of the wide usage of these materials and their adaptability, this study is limited to these classic PZT ceramic material. It is assumed that the physical mechanisms of this material is similar to other PZTs and therefore the results of these experiments can be adapted for other materials. Table. 1 depicts the physical properties of the soft *PZT* compositions used in this study. The specific material used in this study is *P188* obtained from Quartz & Silice, France, Navy type II.

II. BACKGROUND

In the first part of this article, we will briefly discuss the classic dynamic observations and characterizations of a piezo ceramic dielectric behavior, with emphasis on the high and weak excitation time slope dynamic behaviors. The current simulation methods used to produce the dynamic results will be discussed along with frequency dependent hysteresis models, scaling laws, and current aging simulations. The fractional derivative operator and its use for simulating

these dynamic behaviors will be explained, including distinguishing between high and weak excitation time slope dynamic behaviors. The last part of this article will be devoted to the experimental methods for aging and a comparison/validation of the predicted model using experimental results.

TABLE I
MAIN CHARACTERISTICS OF STANDARD P188 CERAMIC GIVEN BY THE MANUFACTURER

Parameters	Symbol	Units	Typical Values
Density	ρ	10^3 kg m^{-3}	7.7
Poisson's constant	σ		0.3
Curie point	Tc	°C	340
Dielectric permittivity	$\varepsilon^T_{33} / \varepsilon_0$		1850
Piezoelectric coefficient	d_{33}	pC/N	425
Dielectric dissipation factor (1 KHz)	$tg\,\delta$	%	≤ 2

III. CLASSIC EXPERIMENTS FOR DYNAMIC DIELECTRIC PROPERTIES - UNDER HIGH SLOPE EXCITATION FIELD STIMULI (dE/dt, dT/dt >> 1)

A. High amplitude levels: Frequency dependence of the dielectric hysteresis

Below the material Curie temperature, at low frequencies (f << 1 Hz) and for high excitation amplitudes, loop-like hysteresis curves can be observed by plotting the spontaneous polarization P, versus electrical field E, or versus an external mechanical stress T. As the excitation frequency increases and exceeds the quasi-static threshold, the piezo ceramic hysteresis cycle starts to show frequency dependence. This dependence can be observed by plotting the frequency variation of the hysteresis area. On a very large frequency bandwidth, this loop area remains constant up to the quasi-static limit, then increases up to a maximum value that depends on the excitation amplitude. For higher frequency levels, the loop area quickly decreases to a value close to zero.

B. Weak amplitude levels: dielectric spectroscopy

Dielectric spectroscopy consists of measuring the dielectric properties of a medium as a function of frequency and under weak amplitude electrical excitation. Dielectric spectroscopy is

based on the interaction of the sample electric dipole moment and the external electrical field E excitation (the dielectric permittivity ε). A ferroelectric ceramic frequency dependence is also called dielectric relaxation and it provides interesting information about the piezo ceramic nature of a material and about the physical relations describing the polarization behaviour. This relaxation is usually described in terms of permittivity as a function of frequency.

IV. CLASSIC EXPERIMENTS FOR DYNAMIC DIELECTRIC PROPERTIES - UNDER WEAK SLOPE EXCITATION FIELD STIMULI (dE/dt, dT/dt << 1)

A. Aging

Ferroelectric ceramics are subject to fatigue due to loads acquired when the materials are used in real world applications along with natural degradation over time, aging. The physical mechanisms of both degradations are similar and linked to pinning defects which are a result of doping and intentionally impurities in the ferroelectric ceramic material. Donor or acceptor dopants, mobile vacancies, and holes are examples of typical defects.

The main consequence of aging is a reduction of the remanent polarization for poled samples and a decrease in the global polarization level during bipolar electric cycling. Effects are amplified for donor-doped materials with electric fields larger than the coercive field [19]. The microscopic origin of aging in a piezo ceramic has been well studied in the scientific literature. The usual physical interpretation of these effects is that mobile charged defects stabilize a given domain configuration thereby generating an internal bias field that counteracts the polarization reversal when an electric field is applied. Three major mechanisms are predominant in the aging process:

_ The volume effect coming from defect dipoles (acceptor ion plus oxygen vacancy) having an electric dipole moment as well as an elastic dipole moment due to the deformation of the unit cell [20][21].

_ The domain effect which can be defined as a migration of charged defects in the direction of the domain walls creating extra pinning defects [22].

_ The interface effect (also called grain boundary effect) which can be explained as the diffusion of ions, vacancies or electrons to grain boundaries creating space-charge layers [23].

V. CLASSIC MODELS AND SIMULATION METHODS OF THE DYNAMIC DIELECTRIC PROPERTIES - UNDER HIGH SLOPE EXCITATION FIELD STIMULI (dE/dt, dT/dt >> 1)

A. High amplitude levels: Frequency dependence dielectric hysteresis models

The simulation of high amplitude excitation dynamic dielectric properties (hysteresis frequency dependence) over a large range of excitation frequencies has been widely studied and documented in the scientific literature [24]. Several methods exist. Two or three-dimensional Ising-type models (multi-scale approach) are used [25][26], but are time intensive and variation is often present between simulated and experimental results. Another method is extension of the quasi-static contribution (Preisach model, Jiles-Atherton model, etc) to dynamic behaviour using the first or a second order additional dynamic contribution provides good results but on a very narrow excitation frequency bandwidth. Neural networks [27] or phase shifting operators [28] exhibit various limitations such as amplitude, frequency, etc and can only be used directly in the field of an associated application.

B. Scaling laws

There are various methods to observe and to model the dynamic dielectric behaviour of a piezo ceramic. One such method is called “scaling laws”, and it consists of focusing on the hysteresis

area variations only [29, 30]. The evolution of the hysteresis area <A> is studied as a function of the excitation amplitude E_0 and frequency f. This method is used to support use in industrial applications [31]. <A> is the energy dissipation in one cycle of domain reversal, and it is related to the non-equilibrium first order phase transitions in ferroelectrics. The hysteresis area provides of interesting information about the dynamics of ferroelectric domains including characteristic times of domain nucleation and domain boundary motion during reversal processes. <A> ∞ f $^{\mu 1}E_0^{\mu 2}$ (where μ1 and μ2 are real parameters depending on the geometry and the nature of the system) is the classic "scaling law" it has been developed as a first attempt to tackle investigations on ultrahigh-frequency ceramics. Frequency exponent of 0.3 to 0.6 were found for classic piezo-ceramics. Scaling laws are an interesting way to understand losses in ferroelectrics but they are not useful simulation tools for modelling ferroelectric aging behaviour as time domain variations cannot be considered.

C. Weak amplitude levels: dielectric spectroscopy simulation

Dielectric spectroscopy is classically described in terms of permittivity as a function of frequency:

$$\varepsilon^*(\omega) = \varepsilon'(\omega) - i\varepsilon''(\omega) = \varepsilon_\infty + \frac{\Delta\varepsilon}{\left(1 + (i\omega\tau)^\alpha\right)^\beta} \quad (1)$$

Here ε_∞ is the sample permittivity under high-frequency excitation, $\Delta\varepsilon = \varepsilon_s - \varepsilon_\infty$ where ε_s is the quasi-static, low-frequency permittivity, and τ is the characteristic relaxation time. ε'(ω) and ε''(ω) are the real and imaginary parts of the permittivity, respectively. For ideal systems α = β = 1, this leads to the Debye complex formula [32]. In alternate cases such as $BaTiO_3$, a classic piezo ceramic β = 1 but α ≠ 1. Materials of this type use the Cole-Cole model [33, 34]. α is linked to the distribution of relaxation time (0 < α < 1). In the case of ferroelectric polymers, optimal results

are obtained with $\alpha \neq 1$ and $\beta \neq 1$. In these cases, the unsymmetrical characteristics and broadness of the dielectric dispersion curve must be considered and can be done using an extension called the Havriliak-Negami [35] relaxation.

VI. CLASSIC MODELS AND SIMULATION METHODS OF THE DYNAMIC DIELECTRIC PROPERTIES - UNDER WEAK SLOPE EXCITATION FIELD STIMULI (dE/dt, dT/dt << 1); AGING SIMULATION

The role of space charges of different natures including electronic, ionic, and, mixed has been pointed out in many studies in the context of aging [20][36]. When comparing aging models, several approaches have been taken including, 1-D, 2-D, and 3-D models. Due to geometric considerations, 1D quantitative models generally vary in comparison to experimental samples. 2-D and 3-D drift diffusion models provided valuable contributions and predictions because they provide information useful to understanding the aging mechanism for both poled and unpoled piezo ceramics. Such models show a time dependent internal bias field characterized by ferroelectric properties including domain width and surface boundary charge, relaxation constant, and temperature dependency [37]. Once this bias field is correctly defined and set for a given sample, the classic $P(E_{eff})$ simulation scheme can be used for simulation of polarization variations, where E_{eff} stands for the effective electric field which is the sum of an external electric field and the time dependent internal bias field.

$$E_{eff}(t) = E_{ext}(t) + E_{bias}(t) \tag{2}$$

VII. FRACTIONAL DERIVATIVE CONTRIBUTION FOR THE SIMULATION OF THE DYNAMIC PROPERTIES - UNDER HIGH SLOPE EXCITATION FIELD STIMULI (dE/dt, dT/dt >> 1)

A. High amplitude excitation

The fractional derivative operators can be solved in both the time and frequency domains. Industrial applications of piezo ceramics under high levels of external excitation like micro-positioning or mechanical vibration energy harvesters usually require real time control and consequently time variations of the ceramic property must be known [5]. For these reasons, the numerical scheme we have been developed is a time domain based solution. It relies on the extension of a hysteresis quasi-static contribution to the dynamic behaviour. As explained previously, the classic dynamic contribution is made from the material's density product to the polarization's first order time derivation. Unfortunately, after comparison with experimental results it is easy to see that a first order consideration can only be used with moderate accuracy on a very narrow frequency bandwidth. To overcome this issue, previous research [14]-[17] has used fractional derivatives to balance the low frequency and the high frequency components in a different way than the limited straight time derivative. To introduce a fractional time derivative $d^{\alpha}B/dt^{\alpha}$ in the system equation, the Grünwald–Letnikov or the Riemman–Liouville definitions can be applied [38]. Both are particular cases of a general fractional order operator named in fractional calculus, the first one represents the α order derivative, while the other one represents the α fold integral. Consequently the class of functions described by the Riemman–Liouville definition is broader (functions must be integratable) than the Grünwald–Letnikov ones. However, for a Grünwald–Letnikov type function, both definitions are similar. In this study, the Riemman-Liouville form with $\alpha \in [0,1]$ has been chosen where $\Gamma(.)$ represents the Euler gamma function.

$$\frac{d^{\alpha} f(t)}{dt^{\alpha}} = D_t^{\alpha} f(t) = \frac{1}{\Gamma(1-\alpha)} \frac{d}{dt} \int_{-\infty}^{t} (t-\tau)^{-\alpha} f(\tau) d\tau \qquad (3)$$

According to equation 3, the fractional derivative of a function f(t) is actually the convolution of f(t) by $t^{\alpha}/\Gamma(1-\alpha)$, where α is the order of the fractional derivation. The additional time derivative present in the formula coincides with the positive occurrence of the gamma function, Γ(.), leading to its convergence towards a finite value. Based on this function, fractional derivatives include memory of the previous states. From a spectral point of view, a powerful consequence of a fractional derivative order is that the frequency spectrum f(ω) of f(t) will be multiplied by $(j\omega)^{\alpha}$ instead of jω for a first order derivation. Consequently, the fractional derivative provides a possibility to satisfy the balance requirements, previously mentioned between it uses the the f(ω) low and high frequency components. As described in [14]-[17], the fractional derivative dynamic term is introduced in the lumped quasi-static contribution and is used as a dynamic contribution (Eq. 5). Fig. 1 gives the block-scheme illustration of eq. 4.

$$\rho . \frac{d^{\alpha} P(t)}{dt^{\alpha}} = E_{dyn}(t) - E_{stat}(P(t)) \qquad (4)$$

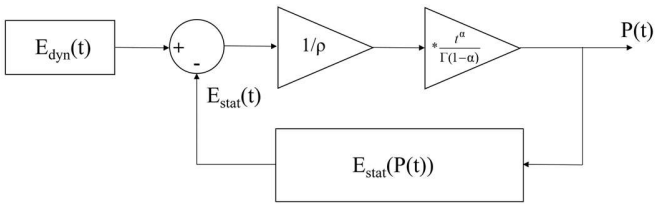


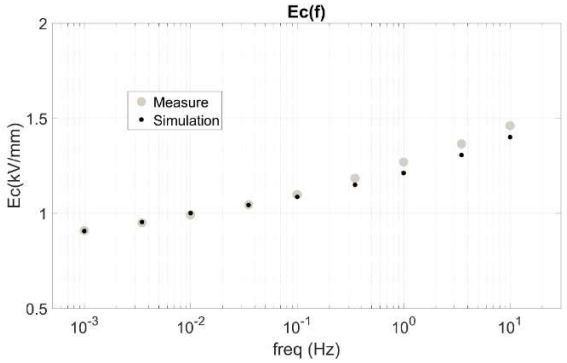
Fig. 1 – Block diagram illustration of the whole dynamic ferroelectric hysteresis model.

In this diagram, P is the polarization field, E_{dyn} represents the electric field excitation, E_{stat} is the quasi-static contribution [6][14]-[17], ρ is a damping constant and α is the fractional order.

Simulated and measured Ec (coercivity), versus f (frequency) variations are shown in Fig. 2 to illustrate the model accuracy. Hysteresis cycle comparisons under fundamental plus harmonic 5 - type waveform are also presented to illustrate the simulation's accurate behaviour under both major and minor hysteresis loop situations. The simulated data and experimental data taken from a piezo material, P188 show agreement in trend and magnitude in relation to hysteresis

loops and coercivity.

A)



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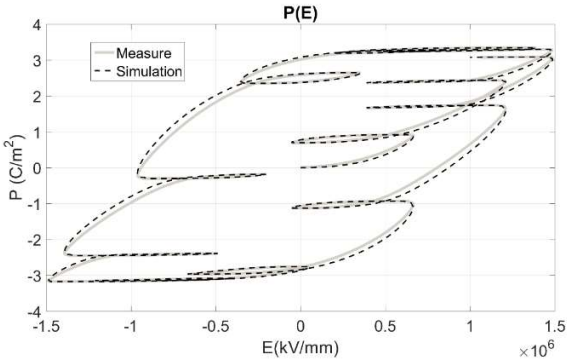

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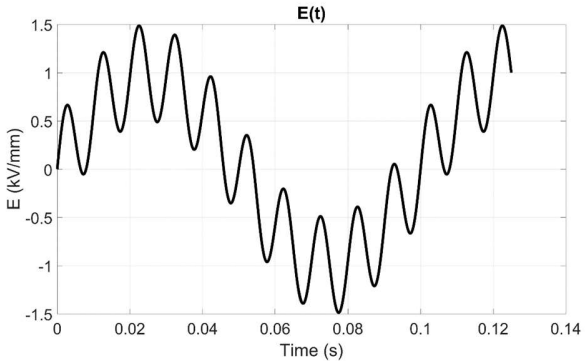


Fig. 2 – A Comparison simulation/measure for the variation of the coercivity Ec versus frequency f, 2 – B Comparison simulation/measure for hysteresis cycle plot under harmonic-type electric field excitation E, 2 – C Time variation of the harmonic type electric field E.

TABLE II
SIMULATION DYNAMIC PARAMETERS

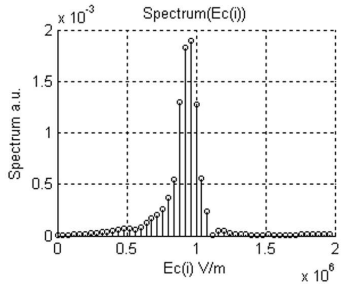
Dynamic parameters	Symbol	Units	Value
Dumping constant	ρ	$V.s^{\alpha}.m.C^{-1}$	$6.\ 10^{4}$
Fractional order	α		0.52

The dynamic parameters we used for this simulation are given in tab. 2.

The quasi-static contribution of this material has been previously described in past work and these values were used for this analysis [14]. The quasi-static parameters (γ, σ, the spectrum distribution function) for the P188 PZT were taken from literature [14] and are listed here in Table. 3.

TABLE III
SIMULATION QUASI-STATIC PARAMETERS

Static parameters	Symbol	Value
Anhysteretic parameter	ϒ	50 000
Anhysteretic parameter	σ	0.3
Spectrum		



B. Under weak amplitude excitation, dielectric spectroscopy simulation

As explained in the dielectric spectroscopy simulation methods section of this article, the Debye equation has been used to simulate the dielectric relaxation response of an ideal, non-interacting population of dipoles with an alternating external electric field E. Unfortunately, for the classic piezo PZT ceramic, the frequency behaviour is far from ideal. The fractional Cole-Cole equation has been utilized to improve the success of simulations. In the Cole-Cole approach, α, the fractional order is linked to the distribution of relaxation time. Assuming E and P harmonic-type waveforms, it is possible to establish a link between the dielectric spectroscopy Cole-Cole model and the fractional derivative high amplitude field consideration described previously

[39][40]. After analysis t of the equation an interesting relation between τ, α, ρ, and ε_s becomes apparent:

$$\tau^{\alpha} = \rho.\varepsilon_s \qquad (5)$$

Comparisons between experimental results using the fractional Cole-Cole equation and simulations on Cole-Cole-type plots for classic PZT ceramic are available in literature [40] and can be used as validation of the fractional used under weak amplitude excitation. Havriliak-Negami advanced theory is sometimes necessary to gain high correlation between experimental and simulated results. Similarly, comparison with high amplitude models can be established but the comparison leads to a series of fractional contributions instead of a single term as in the Cole-Cole equation:

$$\sum \rho_i . \frac{d^{\alpha_i} P}{dt^{\alpha_i}} \qquad (6)$$

In past work [40] comparisons between simulations and experimental results are given for Cole-Cole type plots (i.e. variations of the imaginary versus real part of the permittivity under frequency sweep variations of the excitation field). High correlations between experimental and simulate data provide validation of the method.

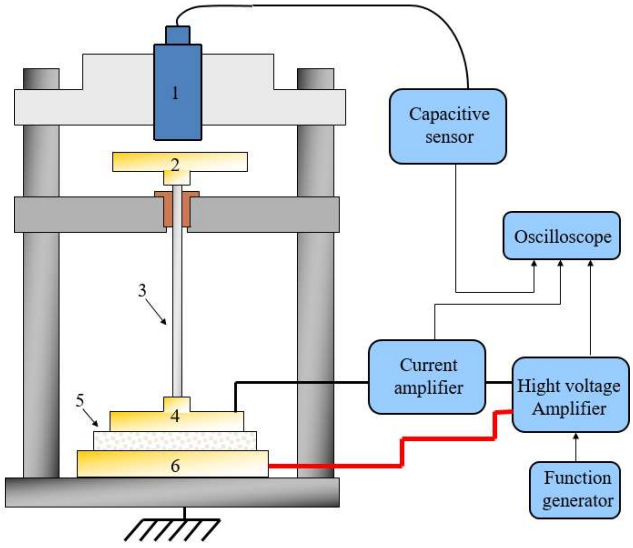
As described and explained in the second part of this article describing current research of simulation methods, the time domain resolution of a fractional derivative includes memory of the previous states. A first order derivation of a f(t) function uses only the f(t-dt) value, whereas the fractional derivation needs the full function f history, which numerically speaking means a series of all the time discretized values of f since the very beginning of the simulation. This property is particularly interesting in our case because it means that even if the electric field excitation becomes null, as it is the case during aging, the polarization value will still evolve

because of the memory of the previous states. The aging simulation is consequently based on eq. 4, where the time window is set to a year. E is nonzero during the poling process and null elsewhere. Once poled, simulated time variations of the remanent polarisation are the direct consequence and manifestation of the piezo ceramic aging.

C. Experimental methods and results

The electric current variations due to depolarizing during aging processes are extremely weak. Such variations are overwhelmed by the surrounding electromagnetic noises and consequently impossible to be measure using standard test methods and laboratory amplifiers. For these reasons, we have used another method to monitor these variations. Remanent polarization of a piezo ceramic after the poling process is useful to monitor for measuring piezo ceramic aging. To measure the remanent polarization, there is no way to avoid a full hysteresis cycle type excitation and consequently a new distribution of the domain which would effectively erase the aging data by resetting the domain. To overcome this issue we have decided to take another experimental approach. As explained, remanent polarization is challenging to monitor during the aging process, but other physical properties such as the permittivity ε or d_{33} the piezo-electric coefficient can be measured without any alteration of the material or aging process. d_{33} is a non-linear but provides a very relevant image of the remanent polarization [9, 10]. By knowing $d_{33}(Pr)$ we can then indirectly follow the Pr evolution during the aging process. To obtain the $d_{33}(Pr)$ characteristic curves, systematic pre-characterisations of all piezo ceramics experimental samples was conducted. In this work, the aging is assumed to correspond to a relaxation of the polarization only, without any degradation or fundamental change in the microstructure, leading to the assumption that the electrostriction is rather constant. Alternate methods exist for

measuring the remanent polarization, but generally require larger number of samples and alternate assumptions must be made using these methods including the same initial remanent polarization state. The test set-up can be seen in fig 3. These pre-characterisations consist of exciting the material with a 1.5 period triangular-type waveform electric field E (fig. 4 – A) in order to reach remanent polarization while increasing the value of the maximum electric field excitation (fig. 4 – B). Once remanent polarization is reached, the d_{33} coefficient is measured. The pre-characterisations leads to the $d_{33}(Pr)$ characteristic (fig. 4 – C) which will be used later as a standard to return the remanent polarisation in the aging experimental process. Cylindrical specimens with diameters of 6.35 mm and heights of 4mm were subjected to the electric field using a Optilas Trek high-voltage supply.



1 - Capacitive sensor

2 - Moving disc

3 - Rod

4 - Moving electrode

5 – Ferroelectric ceramic

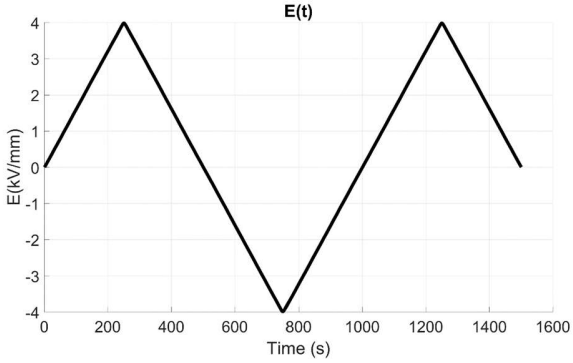
6 - Clamped electrode

Fig. 3 – Pre-Characterisation experimental setup.

We assumed constant temperature and pressure during the process and no additional mechanical constraints were applied to the material. In order to avoid dielectric breakdown, the

specimens were electrically insulated with silicon grease. The electric displacement was calculated by charge measurements using a Kistler amplifier 5011, and the polarization field was calculated by integration of the charge measurements. Fig. 3 gives an illustration of the experimental setup used for all pre-characterizations. To measure, the d_{33} coefficient, a piezometer system from Piezotest Pte Ltd was used.

A)



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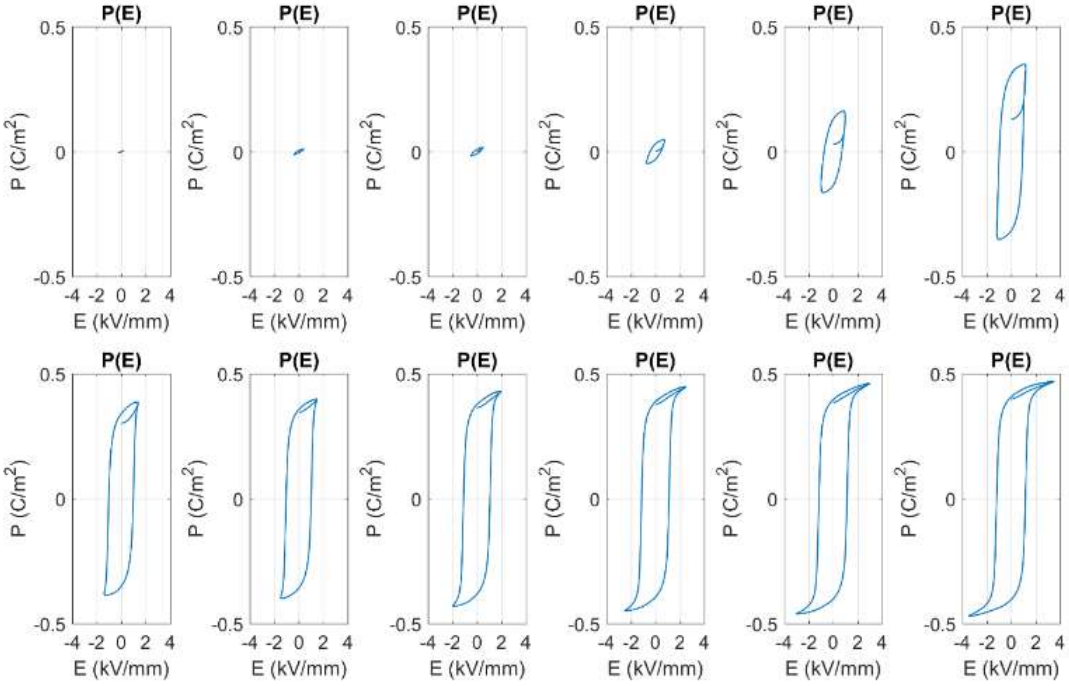

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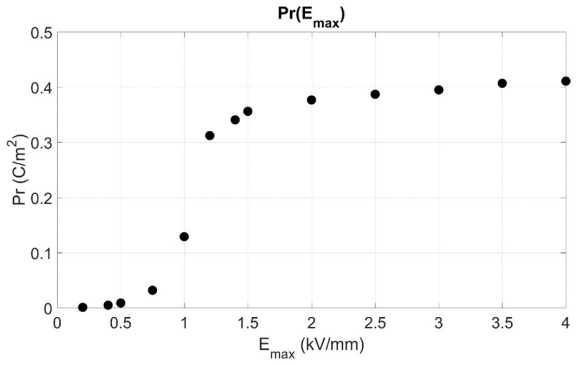

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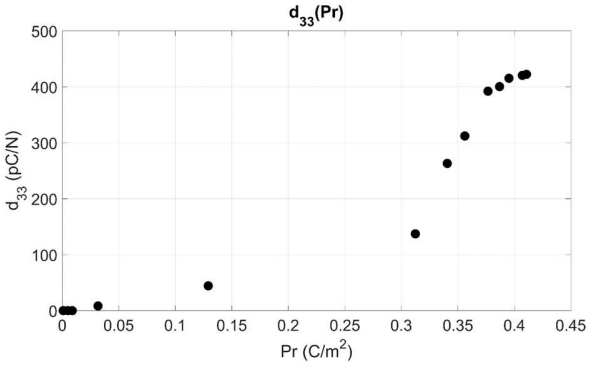


E)

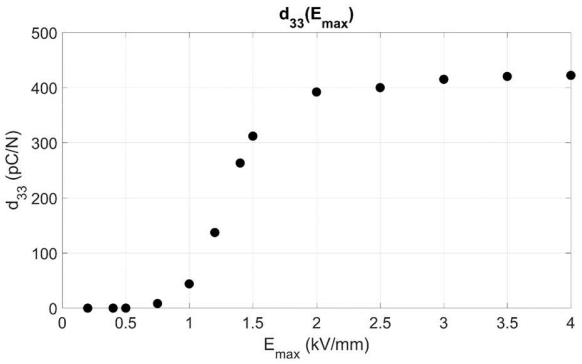


Fig. 4 – A 1.5 period triangular-type waveform electric field *E*, 4 – B related hysteresis cycles, 4 – C *Pr(E)* characteristic curve, 4 – D $d_{33}(Pr)$ characteristic curve, 4 – E $d_{33}(E)$ characteristic curve.

After obtaining baseline values, monitoring the aging process can begin. A final 1,5 period triangular-type waveform, 4kV/mm amplitude electric field E excitation is applied to all test

samples in order to reset the domain distribution and to reach remanent polarization. After that, d_{33} will be monitored regularly over a 200 day duration to observe the aging effects. The first measure is done 75 seconds after polarization. The initial experimental result is depicted in Fig. 5 and it shows the time variation of d_{33}. As envisaged, a quasi-constant negative slope in the logarithmic scale can be observed. It is also noteworthy that a variation occurred at approximately 10^5 seconds. This could be attributed to changes in the laboratory environment or variations in the sample itself.

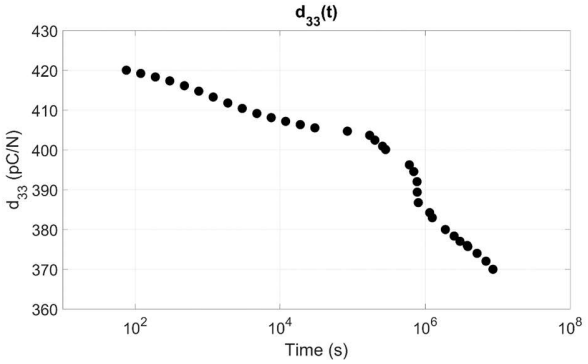


Fig. 5 – $d_{33}(t)$ variations due to aging effect.

Fig. 6 shows the remanent polarisation evolution. To get this curve, a 1D interpolation from $d_{33}(Pr)$ characteristics is performed.

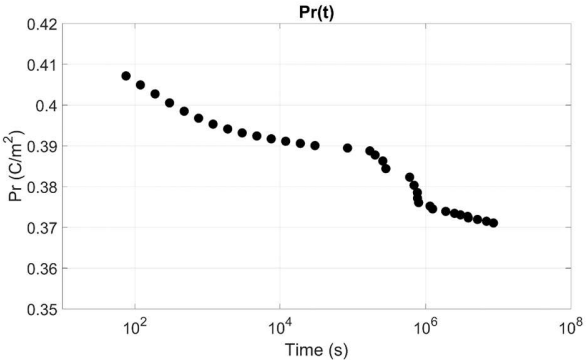


Fig. 6 – Pr(t) variations due to aging effect.

The illustration in fig. 7 shows the *P(E)* simulation results, under both the poling situation (first part of the curve, from *P = 0* to *P = Pr*) and the aging behaviour (slow variation of the remanent polarization).

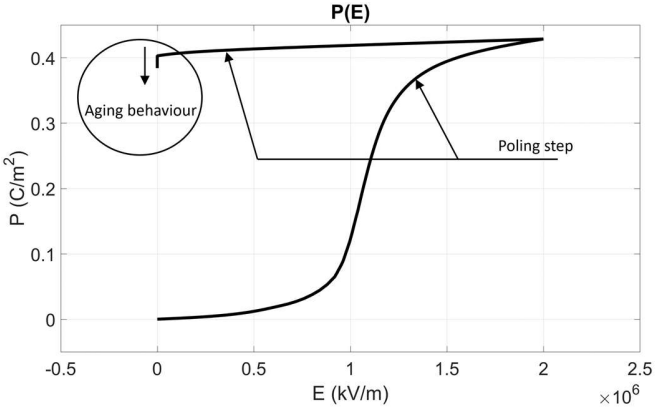
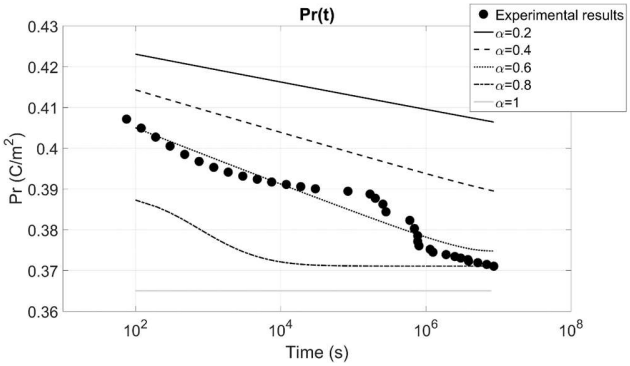


Fig. 7 – P(E) under both poling and aging situation.

Fig. 8 depicts the simulated results and experimental results. Fig 8-A shows Pr(t) variations for different values of α the fractional order ranging from 0.2 to 1. Fig 8- A shows best fit with α = 0.6 when comparing the experimental and simulated results. Fig 8 – B shows a comparison between the experimental Pr(t) and the simulated one for the set of parameters giving the best match (α = 0.6). For all simulation results ρ is maintained constant at ρ = 6 10^4.



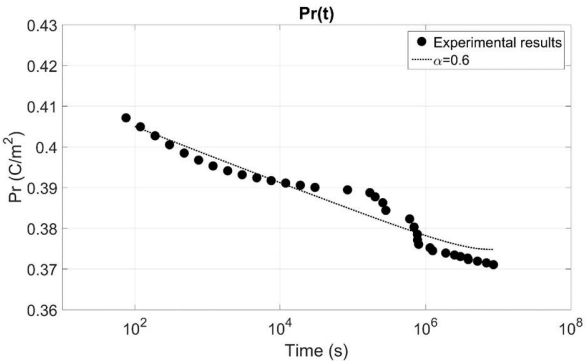


Fig. 8 – Comparison simulations/experimental results for the evolution of the remanent polarization versus time (A for different value of α, B α = 0.6).

It is note-worthy that the best results when comparing simulations to experimental results have been obtained with a value of (α = 0.6) which is relatively close to 0.52 corresponding to the fractional order leading to the best matches under *P(E)* situations.

In our simulation, a fractional derivative order equal to 1 means a quasi-instantaneous dynamic contribution which will stop right after the electric field excitation removal. This comes from the numerical definition of the first order derivative which to be solved relies on the previous time step. Now for lower fractional order, the resolution needs a window of previous time steps to be solved and the lower the coefficient is, the higher the influence of the initial time steps. From the aging point of view it means that the lower α is, the lower the aging rate will be. An interesting observation is that no matter what the order of the fractional derivative is, if the simulation time is long enough the polarization field will always reach a steady state where no more variations can be observed. The value of α is inversely proportional to this transient phase time.

In Figure 9, the aging rates versus time have been plotted on a linear time scale comparing the simulated and experimental results. Since the aging rate stabilizes after 10^4 seconds, an alternate method of looking at this property was used. In Figure 10, aging versus time decades is displayed.

The experimental aging rate is noisy with an average value around 1.7%. Even if none of the simulation results reproduces strictly the experimental ones, α = 0.6 gives an average aging rate (1.98 %) very close to the experimental one (1.72 %).

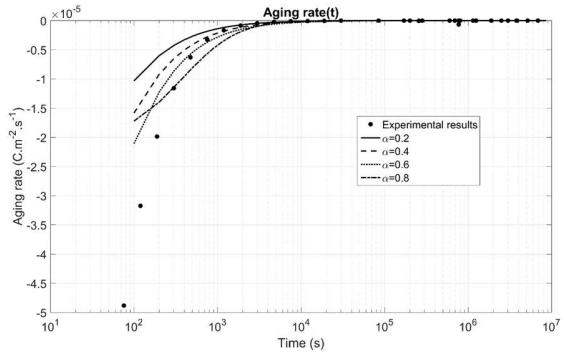


Fig. 9 – Linear Time Scale Aging Rates Experimental Versus Simulated.

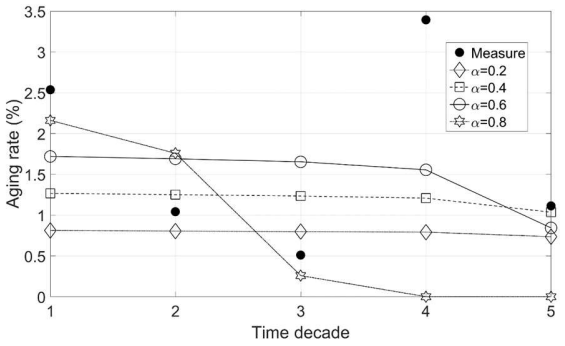


Fig. 10 – Aging Rates versus Time Decades (Decade 1 corresponds to 100 – 1000 seconds.

VIII. CONCLUSION

Dynamic behaviours in ferroelectric materials can be physically explained as a complex combinations of diffusive and dissipative behaviours. The dissipative behaviours are linked to the dynamics of domain wall motion. The diffusive behaviours are linked to mechanical and thermal relaxation. In the past, fractional derivatives have been used with success for the simulation of systems exhibiting dynamic behavior known as anomalous diffusion [41]-[43]. Where first or second order derivation are used in classic diffusion equations, anomalous diffusions are more

complex and consequently require alternative mathematical operators to be simulated. It is also used to complex visco-elastic behavior [44] as an intermediate state between elastic and viscous bodies. The dynamic behavior including the frequency dependence of hysteresis and aging of piezo ceramics is one of those. As discussed in IV-A aging is due to three major mechanisms: the volume effect, the domain effect and the interface effect. These mechanisms can be combined. All together they lead to a macroscopic behavior which can be assimilated to an anomalous diffusion and simulated using fractional derivative operators. In our simulation itself, we have no way to distinguish the aging mechanisms as the model is a phenomenological lump approach with no direct connection to the ferroelectric sample micro-structure. With this non-predictive but phenomenological approach, we focus on reproducing experimental signals as they are measured, physical interpretations can be made in term of anomalous diffusion but because of the space discretization absence there is no local information and no direct connection to the ceramic micro-structure. There is therefore no way to estimate the activation energy responsible for aging such as to differentiate the contribution of charges to the ionic space charges.

It has already been demonstrated that the frequency dependence of the dielectric properties can be correctly modelled in the time domain thanks to fractional derivative operators on a very large frequency bandwidth of external excitations. Up to now, the lowest frequencies have been limited to the hysteresis cycle quasi-static threshold generally in the range of 10 mHz for classic PZT ceramics. In this article, a simulation technique using the fractional derivative dynamic contribution was validated under a wide frequency range and particularly in terms of long time variations like those experience during aging. The simulation was validated using experimental data from a piezo ceramic material, P188. The fractional parameters from the simulation are

conserved through the frequency dependence of dynamic hysteresis cycle and long-time variation of the remanent polarization which can be physically interpreted as a conservation of the physical behaviour even if these parameters are evolving in different scales of time. This work confirms that fractional derivative based simulation is capable of predicting aging in piezo ceramic materials.

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