



Experimental evidence that electrical fatigue failure obeys a generalized Coffin–Manson law



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ABSTRACT

The empirical Coffin–Manson law has been used to characterize the low-cycle mechanical fatigue failure of metallic materials for decades. Our experimental studies reported in this letter have shown that the electrical fatigue failure in dielectrics can be well described by a fitting function having the same mathematical expression as that of the Coffin–Manson law. This observation indicates that the physical mechanism beneath the formation and evolution of atomic disordered structures, the key factor influencing both mechanical and electrical fatigue, might be the same.

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1. Introduction

Electrical fatigue or polarization fatigue phenomena in ferroelectric materials have been extensively investigated in the past decades. However, a general understanding of the origin of such dielectric deterioration is still missing [1,2]. In this letter, our experimental studies on polarization fatigue failure of poly(vinylidene fluoride-trifluoroethylene) [P(VDF-TrFE) 75/25 mol%] copolymer films are reported. The objective of our investigation is to attempt to tackle the polarization fatigue problem from a different perspective and gain a better understanding of it. To be specific, we would like to test whether electrical fatigue can be described by a certain mathematical formula of mechanical fatigue law or not. Our approach is based on the reasoning and argument given below.

When we apply an external voltage to a piece of insulating material, it will undergo volume deformation due to electromechanical coupling effects (electrostriction and flexoelectricity for all dielectrics and plus piezoelectricity for ferroelectric materials). If the applied voltage is a periodic signal with a long duration (electrical cyclic loading), the deformation of the dielectric will become a cyclic process. Thus, the electrical cyclic loading for dielectrics is equivalent to the mechanical cyclic loading for metallic materials. Certainly, we would conjecture that, from the viewpoint of equilib-

rium thermodynamics, there must be some resemblances between mechanical and electrical fatigue.

Furthermore, if the above mentioned deformation remains in the elastic range, there is a pair of balanced forces, $\nabla_r W_e = \nabla_r W_m$, inside the dielectric; here W_e is the electrostatic energy stored in the dielectric, ∇_r is the mathematical symbol representing the gradient with respect to the direction r , and W_m is the mechanical energy, due to the induced deformation, stored in the dielectric. In order to prevent partial discharge channels or, more generally, electrical-breakdown structure precursors, which would eventually lead to the occurrence of electrical breakdown in the dielectric, from initiating and growing, the following force inequality must be satisfied [3]: $\nabla_r W_e \leq \nabla_r W_{max}$; here W_{max} represents the maximum mechanical energy allowed to be stored beyond which electrical-breakdown structure precursors will initiate and grow. In this case, the initiating and growing process of electrical-breakdown structure precursors can be regarded as that of microcracks in mechanical fatigue failure. Thus, there must also be some resemblances between mechanical and electrical fatigue failure.

In this letter, we attempt to test if the electrical fatigue failure in dielectrics can be fitted by using a function that has the same mathematical expression as that of the Coffin–Manson law. Before introducing our fitting function, we first write down the Coffin–Manson law as follows.

The low-cycle fatigue of metallic materials is described by the Coffin–Manson law [4,5], which is given below.

$$\frac{\epsilon_p}{2} \approx \epsilon_f (2N_f)^{-\beta_{CM}}, \quad (1)$$

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here ϵ_p is the induced plastic strain; ϵ_f is an empirical value known as the fatigue ductility coefficient that represents the failure strain for a single reversal; N_f is the number of reversals to failure (life cycles) and β_{CM} is called the Coffin–Manson exponent.

Now we substitute $\frac{\epsilon_p}{2}$ in the above equation with P_r and $\epsilon_f(2)^{-\beta_{CM}}$ with P_f ; then we have the following fitting function

$$P_r \approx P_f(N_f)^{-\beta_{CM}}, \quad (2)$$

where P_r is the remnant polarization; P_f is defined as the electrical failure coefficient that is the failure polarization (the remnant polarization just before electrical breakdown occurs) for a single reversal. For simplicity we define Eq. (2) as a generalized Coffin–Manson law.

To show the potential merit of our studies, it might be necessary to consider and answer two questions before we present our experimental results. The first question is why we choose the Coffin–Manson law as a template for proposing Eq. (2) to describe electrical fatigue failure? Our consideration is that the correctness of the Coffin–Manson law has been experimentally verified during the past six decades, and moreover, the Coffin–Manson law is an empirical law and its physical mechanism is still not completely clear. Therefore, if we could experimentally verify that electrical fatigue failure in dielectrics can be fitted by Eq. (2), then our studies may provide a different route where not only can some experimental and analytical techniques in mechanical fatigue field be borrowed to investigate electrical fatigue but also the similarity between these two fatigue behavior might reveal that their physical origins at the atomic level could be the same. The second question is why we consider polarization fatigue of ferroelectric materials in our studies? In low-cycle fatigue of metallic materials where the Coffin–Manson law is applicable, the stress – strain relationship of the studied metallic sample is no longer linear (plastic deformation occurs). Similarly, in polarization fatigue of ferroelectric materials, the electric field – polarization relationship of the studied dielectric sample is also nonlinear (hysteresis loop appears). Therefore, it would be reasonable to compare polarization fatigue failure data to the fitting curve given by Eq. (2).

In the following sections, our material preparation and experiment procedures are introduced; then follow discussions of experimental results and concluding remarks.

2. Material preparation and experimental procedures

P(VDF-TrFE) 75/25 mol% copolymer films were used in our studies. Its powder was provided by Solvay Solexis and the procedure of preparing P(VDF-TrFE) films is briefly summarized as follows (our procedure is a modified version of the one reported in Ref. [6]): (1) Dissolving P(VDF-TrFE) powder in a butanone solvent; the weight ratio of the powder to the solvent is 12:100; (2) Stirring the P(VDF-TrFE)-butanone solution by using a magnetic stir bar on the surface of a hot plate at 90°C degrees for 1 hour; (3) Spin-coating a thin layer of the solution on the top of a Pt/Ti/SiO₂/Si(100) wafer, previously cleaned with alcohol/DI water, by using a Laurell spin coater. The coating time and the coating rate (RPM) vary in different situations and were used to control the thickness of the layer; (4) Placing the spin-coated wafer in a pre-heated oven at 150°C degrees for 8 minutes; (5) Then turning off the power to the oven and letting the wafer cool down to room temperature naturally.

The fabricated film is shown in Fig. 1. The thickness of the film was measured and its average value is $\sim 3\mu\text{m}$. The Pt layer of the wafer was used as the ground electrode and top electrodes were fabricated on the surface of the film; each of top electrodes is a solid circle, covered by silver paste (PELCO colloidal silver paste from Ted Pella), with a radius of $\sim 2\text{mm}$; there is an appropriate



Fig. 1. The P(VDF-TrFE) copolymer film coated on a Pt/Ti/SiO₂/Si(100) substrate.

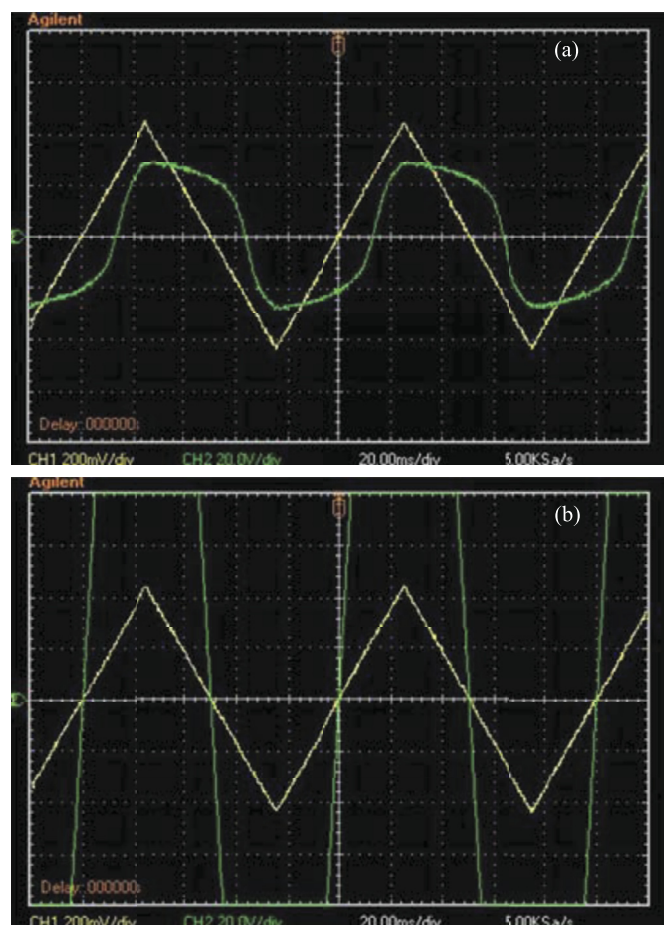


Fig. 2. The waveforms collected from the oscilloscope connected to a standard Sawyer–Tower circuit: (a) the triangle waveform is the input voltage signal and the distorted waveform represents the voltage measured across the reference capacitor of the Sawyer–Tower circuit. In this case, electrical breakdown inside the film has not yet occurred; (b) the triangle waveform is still the input voltage signal but the voltage measured across the reference capacitor jumps beyond the voltage limit of the oscilloscope, which means that electrical breakdown has occurred.

space to separate one top electrode from others so that the occurrence of electrical breakdown at that spot will not affect later measurement at other spots.

Polarization fatigue phenomena of our P(VDF-TrFE) film were studied by using a standard Sawyer–Tower circuit. The input and output waveforms of this Sawyer–Tower circuit are shown in Fig. 2; the triangle waveform is provided by a function generator (Stanford Research Systems DS345) and this signal is also ap-

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