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CREEP-FATIGUE RELATIONSHIPS IN ELECTROACTWE POLYMER SYSTEMS AND PREDICTED EFFECTS IN AN ACTUATOR DESIGN

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Abstract

The paper concerns the time-dependent behavior of electroactive polymers (EAP) and their use in advanced intelligent structures for space exploration. Innovative actuator design for low weight and low power valves required in small plants planned for use on the moon for chemical analysis is discussed. It is shown that in-depth understanding of cyclic loading effects observed through accelerated creep rates due to creep-fatigue interaction in polymers is critical in terms of proper functioning of EAP based actuator devices. In the paper, an overview of experimental results concerning the creep properties and cyclic creep response of a thin film piezoelectric polymer polyvinylidene fluoride (PVDF) is presented. The development of a constitutive creep-fatigue interaction model to predict the durability and service life of electroactive polymers is discussed. A novel method is proposed to predict damage accumulation and fatigue life of polymers under cyclic loading conditions in the presence of creep. The study provides a basis for ongoing research initiatives at the NASA Kennedy Space Center in the pursuit of new technologies using EAP as active elements for lunar exploration systems.

Keywords: Electroactive Polymers, Actuators, Creep, Fatigue, Viscoelasticity, Damage.

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Introduction

In the past decades electroactive polymers (EAP) have been increasingly used in many technological fields including aerospace, transportation, telecommunication, photonics, and bioengineering due to their unique ability to sense and react to external stimuli in the form of electrical signals or mechanical deformations. Polymer based transducers, sensors and actuators have shaped an impressive array of advanced technologies including acoustic microscopy, active vibration control, damage detection in fibrous composites, optoelectronics, and marine fouling prevention. Increasingly, electroactive polymer systems have been used for space exploration in weight sensitive applications such as shape and position control of compliant structures, smart skins, solar sails, deployable membrane mirrors, atmospheric balloons, antennae, reflectors, flexible robotic manipulators for grasping and locomotion, pumps, valves, relays and other applications which have traditionally relied on electromagnetic components. The use of electro active polymers provides numerous advantages in terms of reduced weight, large motions without gearing, and built in position sensing capabilities [1, 2].

The National Aeronautics and Space Administration is currently pursuing a lunar exploration program that will benefit from innovations in system components designed to reduce weight and power consumption without sacrificing reliability. For example, low weight and low power fluid handling valves will be required in small chemical plants planned for use on the moon for chemical analysis. Available valves based on solenoids can produce millions of opening and closing cycles so that valve seat design rather than the electromagnets becomes the limiting life design factor. In these applications, electroactive polymer actuators are considered to be a logical substitute for current electromagnetic devices.

The effective use of electroactive polymers as active elements in actuator design requires

precise understanding of their properties. Many important issues in this regard arise in connection with material nonlinearities, time-dependent effects, and energy losses that characterize the electromechanical response of EAP [3-7]. It is well known that under sustained loading conditions polymers exhibit creep at room temperature with accelerated creep rates at elevated temperatures. The creep behavior of polymers in response to static loads changes dramatically in response to cyclic load histories involving stress reversals or cyclic temperatures. Under such conditions polymers develop much higher creep rates than those observed in static experiments. It is considered that cyclic creep acceleration in polymers is caused by material damage due to fatigue which results in creep-fatigue interaction [8, 9].

Enhanced understanding and accurate prediction of creep-fatigue interaction effects on the performance and longevity of EAP devices is critical. Since such devices are designed to operate in dynamic environments, accelerated material degradation processes are likely to impair the functional performance of EAP systems, similarly to the effects of microcracking that tend to decrease the accuracy of piezoelectric sensors and actuators by weakening the electromechanical couplings in material properties [10].

Experimental Results

A representative group of EAP comprises piezoelectric polymer systems including polyvinylidene fluoride (PVDF), PVDF co-polymers and piezoelectric polymer based composites. At present, this group has reached a greater degree of maturity in terms of technological applications as compared with other electroactive polymers. Essentially, PVDF and PVDF co-polymers represent the principal commercially available high-performance piezoelectric polymers currently in use. These materials are characterized by many attractive properties such as stable response characteristics in a wide frequency range of up to 10^9 Hz, low

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acoustic impedance, high degree of resistance to impact, resistance to moisture absorption, and marginal sensitivity to ultraviolet and nuclear radiation [4-6].

Strong piezoelectric effects in PVDF are obtained through a technological process that involves stretching and polling of extruded thin sheets of the polymer. Stretching provides an alignment of molecular chains in the stretch direction. An applied electric field of 100 kV/mm at an elevated temperature of about 103°C causes permanent polarization which is maintained after the material is cooled to room temperature. These processing and fabrication conditions play a decisive role in defining the electromechanical properties of piezoelectric polymers.

The time-dependent mechanical properties of PVDF have been studied based on a series of quasi-static creep tests and dynamic mechanical tests of 28 μ m thick commercially produced PVDF thin films. In the experiments, PVDF samples have been tested in two in-plane material directions, i.e., parallel (direction 1) and perpendicular (direction 2) to the aligned molecular chains of the polymer. Creep experiments have been performed at 10 different stress levels under sustained loading conditions at room temperature. Strain measurements have been taken by a linear variable differential transformer (LVDT). The creep behavior of PVDF has been also studied using the method of dynamic mechanical testing and analysis. The results of these experiments are illustrated in Figs. 1 and 2 that demonstrate the creep response of PVDF in terms of strain ε (%) as a function of time t [11-12].

It has been determined that at stresses σ below 57% of the yield stress $\sigma_{Y1} = 45 \text{ MPa}$ in the material direction 1, and below 76% of the yield stress $\sigma_{Y2} = 39$ MPa in the direction 2 the creep response of PVDF is linear such that the strain histories $\varepsilon(t)$ obtained at various constant stresses and normalized based on the respective stress can be represented by a single curve

known as creep compliance. The creep compliances for PVDF, $C_n(t) = \varepsilon(t)/\sigma$ (n= 1, 2), for material directions 1 and 2 are shown in Figs. 3 and 4, respectively.

Fig. 1. Creep Strain of PVDF (direction 1).

Fig. 2. Creep Strain of PVDF (direction 2).

Fig. 3. Creep Compliance of PVDF (direction 1).

Fig. 4. Creep Compliance of PVDF (direction 2).

Cyclic creep response of PVDF has been studied in the stretch direction of the polymer (direction 1) because of higher creep rates as compared with the direction 2. The experimental investigation involved a series of tests under the conditions of superimposed static and cyclic loads, i.e., $\sigma(t) = \sigma_m + \sigma_a \sin \omega t$ applied in asymmetric tension-tension mode. The applied cyclic amplitude, σ_a was much smaller than the mean stress σ_m , i.e. $\sigma_a \ll \sigma_m$. The results were obtained in terms of creep strains ε(t) and interpreted in terms of normalized creep strain ε(t)/σ_m. At least three -

five identical experiments were repeated under the same loading conditions to ensure reproducible results [9, 13].

This study has demonstrated that under cyclic loading conditions creep rates of PVDF accelerated with an increase of the amplitude and frequency of the cyclic load. These effects are illustrated in Fig. *5* which provides a comparison between cyclic creep strains c(t) normalized by the mean stress σ_m and the respective creep compliances obtained under static loading conditions. Cyclic creep of PVDF depicted by curve 3 has been obtained for the following loading conditions: $\sigma_m = (0.45)\sigma_Y$, $\sigma_a = (0.014)\sigma_Y$, and $\omega = 10$ Hz. The curve 4 was obtained for $\sigma_m = (0.34)\sigma_Y$, σ_a = (0.068) σ_y , and ω = 5 Hz. The curves 1 and 2 represent static creep compliances obtained at σ = (0. 34) σ _Y and σ = (0. 45) σ _Y, respectively.

Fig. *5.* Cyclic and Static Creep Responses of PVDF.

These and other results reported in [9, 13] demonstrate that piezoelectric polymers undergo measurable creep acceleration when subjected to superimposed static and cyclic loads.

Consistently, cyclic creep rates tended to increase with an increase of frequencies or amplitudes. This phenomenon can be explained by the presence of several interactive mechanisms: creep, hysteretic heating, and fatigue damage. The contribution of each individual mechanism depends on the loading and temperature conditions. In particular, experiments indicate that fatigue failure of polymers is dominated by hysteretic heating at higher stress levels and frequencies, whereas damage accumulation processes become critical at lower stresses and frequencies [8, 14]. In general, cyclic damage evolution in polymers involves several consecutive stages, the formation of crazes, craze growth, crack nucleation and crack propagation [14]. Even before the appearance of crazes, polymers subjected to cyclic loading regimes tend to undergo measurable changes of their density, shear modulus, and internal damping. The study reported in [15] has shown direct correlation between the sensitivity of polymers to cyclic frequency effects and damage evolution in terms of nucleation and propagation of crazes. These observations, however, have not been consistently characterized or quantified. To date, the creep-fatigue interaction effects in the behavior of electroactive polymers remain unexplored.

Modeling of Static Creep

As discussed in the foregoing section, within the range of stresses not exceeding *25* MPa the creep properties of PVDF can be represented by creep compliances shown in Figs. 3 and 4. Respectively, the polymer can be characterized by the constitutive equations of linear viscoelasticity based on the Boltzmann superposition principle which states that since strain is a linear function of stress the total effect of applying several stresses is the sum of the effect of applying each one separately. The application of this principle leads to the stress-strain relation in the form [16, 17]

$$
\epsilon(t) = \sigma(0) C_{n}(t) + \int_{0}^{t} C_{n}(t-\tau) \frac{d\sigma}{d\tau} d\tau,
$$

(1)

where the stress σ and strain ε are functions of time t, and C_n (n = 1, 2) denote the creep compliances in the respective material direction of PVDF.

The creep compliances $C_n(t)$ of PVDF can be represented analytically in the form

$$
C_n(t) = a_n + b_n(t_n^{\alpha}), \quad n = 1, 2
$$
 (2)

where the coefficients a_n , b_n and α_n are defined as follows:

Direction 1:
$$
a_1 = 3.206 \times 10^{-10}
$$
; $b_1 = 5.018 \times 10^{-11}$; $\alpha_1 = 0.107$ (3)

Direction 2:
$$
a_2 = 3.514 \times 10^{-10}
$$
; $b_2 = 0.111 \times 10^{-11}$; $\alpha_2 = 0.085$ (4)

Substitution of Eq. (2) into Eq. (1) leads to an integral equation which can be solved

using the Laplace transformation method in terms of the stress function $\sigma(t)$ in the form

$$
\sigma(t) = \frac{\varepsilon(t)}{a_n} - \frac{1}{a_n} \int_{0}^{t} \sum_{n=1}^{\infty} \frac{(b_n \alpha_n / a_n) \Gamma(\alpha_n) (t - \tau)^{\alpha_n}}{(t - \tau) \Gamma(n\alpha_n)} \varepsilon(\tau) d\tau ; n = 1, 2
$$
 (5)

where Γ (α_n) and Γ ($n\alpha_n$) are gamma functions, and the coefficients a_n , b_n , and α_n ($n = 1,2$) are defined by Eqs. (3 and 4).

In the case of constant strain $\varepsilon(t) = \varepsilon_0 = \text{const.}$, Eq. (5) is of the form

$$
\sigma(t)/\varepsilon_{o} = \frac{1}{a_{n}} [1 - \int_{0}^{t} \sum_{n=1}^{\infty} \frac{(b_{n} a_{n}/a_{n}) \Gamma(\alpha_{n})(t-\tau)^{\alpha_{n}}}{(t-\tau) \Gamma(n\alpha_{n})} d\tau] = \frac{1}{a_{n}} [1 - R_{n}(t)] ; n = 1, 2
$$
 (6)

where

$$
R_n(t) = \int_0^t \sum_{n=1}^{\infty} \frac{(b_n \alpha_n / a_n) \Gamma(\alpha_n) (t - \tau)^{\alpha_n}}{(t - \tau) \Gamma(n\alpha_n)} d\tau ; n = 1, 2
$$
 (7)

denote the relaxation functions of PVDF in both in-plane material directions. The set of Eqs. (1-7) completes the characterization of PVDF in the linear viscoelastic range of creep properties under static loading conditions.

Modeling of Creep-Fatigue Interaction

To characterize creep-fatigue interaction effects in PVDF, a constitutive model has been developed based on the principles of linear viscoelasticity and continuum damage mechanics [16-19]. The concept of a fictitious undamaged continuum with effective material characteristics has been used to represent fatigue damage in the material. Specifically, damage evolution in PVDF has been characterized by a damage function D which depends on the number of cycles N, i.e., $D = D(N)$. The damage function is defined within the limits $0 \le D \le D^*$, where the low limit represents undamaged material, $D(0) = 0$, and D^* characterizes the degree of damage at which the response of the material cannot be treated anymore as linearly viscoelastic. The value of D* must be determined experimentally.

Based on these assumptions, the properties of a fatigue damaged linear viscoelastic material can be defined by the following constitutive equation

$$
\varepsilon_{\rm D}(t, N) = C_{\rm eff}(t, N)\sigma(0) + \int_{0}^{t} C_{\rm eff}(t-\tau, N)\frac{d\sigma}{d\tau}d\tau
$$
 (8)

where

$$
C_{\text{eff}}(t, N) = C(t)/[1 - D(N)]
$$
\n(9)

denotes the effective creep compliance of the material with damage and C(t) is the creep compliance of undamaged material. Note that, at $N = 0$, $C_{\text{eff}}(t, 0) = C(t)$. In Eq. (8), $\varepsilon_D(t, N)$) denotes the strain in the damaged material produced in response to the stress history $\sigma(t)$.

It follows from Eq. (9) that the damage function $D(N)$ can be represented in the form

$$
D(N) = 1 - \frac{C(t)}{C_{\text{eff}}(t, N)}
$$
(10)

Based on this representation an experimental protocol can be developed to characterize the damage function $D(N)$ since the creep compliances $C_{\text{eff}}(t, N)$ and $C(t)$ can be determined experimentally. Specifically, the creep compliance C(t) can be obtained from static creep experiments at a constant stress σ_0 by normalizing creep deformations based on the applied stress. The same method can be applied to a damaged material after N number of cycles, providing the effective creep compliance $C_{\text{eff}}(t, N)$. Note that this approach is based on the assumption that the damaged material is linearly viscoelastic.

Since the creep compliances $C(t)$ and $C_{\text{eff}}(t, N)$ are obtained by normalizing the experimentally determined strains in the damaged and undamaged materials, $\varepsilon_D(t, N)$ and $\varepsilon(t)$, respectively, such that

$$
C_{\rm eff}(t, N) = \varepsilon_D(t, N)/\sigma_o \text{ and } C(t) = \varepsilon(t)/\sigma_o \tag{11}
$$

substitution of Eqs. (11) into Eq. (10) provides an alternative equation for computing the damage function

$$
D(N) = 1 - \frac{\varepsilon(t)}{\varepsilon_D(t, N)}
$$
(12)

The proposed creep-fatigue interaction model has been validated experimentally. PVDF samples were prepared from commercially available thin sheets of the material with deposited silver electrode layers on both surfaces of the polymer. The total thickness of the samples including the thickness of silver layers was 46 μ m, with the thickness of the PVDF layer of 28

m. The samples had in-plane dimensions 22 mm x 1.5 mm, and were tested in the direction of the aligned molecular chains of the polymer (direction 1). Tests were performed using a TA Instruments dynamic mechanical analyzer DMA 2980.

As the first step, the creep compliance $C(t) = \varepsilon(t)/\sigma_0$ of undamaged material was. determined from creep tests under sustained loading conditions at the stress level σ_0 = 10 MPa, which is below the viscoelastic linearity limit. The results of these tests are shown by the curve 1 in Fig. 6.

Creep Compliance (1/Pa)x10⁻⁶

Fig. 6. Creep of PVDF (1: Static; 2: Cyclic ; 3: Two-Stage Stress History).

The second series of experiments was conducted under the conditions of superimposed static stress $\sigma_0 = 10$ MPa and cyclic stress with the amplitude $\sigma_a = 4.5$ MPa and frequency ω = 20 Hz. Note that the total maximum stress σ_{max} = 14.5 MPa did not exceed the viscoelastic linearity limit of PVDF determined as 25.65 MPa. The respective cyclic strains were measured and normalized with respect to σ_0 as represented by the curve 2 in Fig. 6. As expected, at this stage, the material demonstrated visible creep acceleration as compared with static creep.

The third series of experiments consisted of two types of tests. Firstly, cyclic damage was generated for over six hours under the same stress conditions as applied at the second stage of the program. After that period only the static load was maintained. The respective effective creep compliances C_{eff}(t) was determined by normalizing the measured creep strain of the damaged material by the respective static stress. The results of these experiments are illustrated by the curve 3 in Fig. 6.

It is clear that under static loading conditions the damage function $D_1 = 0$. Based on the experimental data obtained from the second and third series of tests, the respective damage functions D_2 and D_3 shown in Fig. 7 were determined using Eq. (10).

Fig. 7. Damage Characterization of PVDF.

It follows from these diagrams that cyclic damage in PVDF initiates immediately upon the load application. Both functions D_2 and D_3 increase with the number of cycles N which

characterizes the rate of damage evolution. The damage function D_3 remains constant, which indicates no further damage development in the material in the absence of the cyclic load.

Challenges

Integration of electroactive polymers into structural design provides the capability of controlling the mechanical characteristics of structures in terms of stiffness or damping, or modify the structural response in terms of position or velocity. This type of built-in structural intelligence has been particularly effective in space applications. In particular, the use of EAP in machine components and devices for lunar exploration has been actively pursued at the NASA Kennedy Space Center. In November of 2008, NASA performed a series of experiments at the lunar analog site on Mauna Kea, Hawaii, chiefly to demonstrate the technologies capable of analyzing and extracting oxygen from lunar soil. Experiments like the Regolith Volatiles Characterization (RVC) and the Lunar Water Resource Demonstrator (LWRD) were essentially miniaturized chemical plants for detecting soil volatiles and analyzing water [20]. The plants contained rotary valves, solenoid valves and pumps for routing gases. Ambitious goals have been set for the design of these devices in terms of mass, thermal, and power requirements. In this regard, the existing technologies appear ineffective. It is expected that a promising alternative to traditional design will be provided by electroactive polymer systems that will enable new technological developments for lunar exploration.

It is clear that, in practice, successful implementation of EAP systems directly depends on the degree of understanding of their behavior and properties. To date, considerable progress has been made in this subject area [2, 4]. However, many aspects of the performance and properties of electroactive polymers remain unexplored. In particular, challenges arise due to the sensitivity of polymers to fabrication and temperature conditions, time-dependent effects,

and material nonlinearities. An immediately obvious challenge to fielding electroactive polymers is a lack of understanding of their failure modes, reliability, and the change of electromechanical properties depending on time and temperature. Questions as to the nature of long term creep and fatigue-fatigue interaction effects in polymers must be understood and quantified before EAP can be widely accepted as viable competitors to the electromagnetic actuator technologies currently in use.

Work-in-Progress

To investigate the potential use of EAP in lunar exploration, the Kennedy Space Center is sponsoring research on the creation of simulation models to assist in the design of EAP based active control systems. To illustrate the importance of understanding long term creep and fatigue effects in an EAP element, consider the design of a valve currently under investigation at Kennedy Space Center. A simplified valve schematic and the corresponding valve seal free body diagram demonstrating the acting forces are shown in Fig. 8.

Fig. 8. Valve Schematic and Free Body Diagram.

In this normally open design, a valve seal moves downward on application of voltage to mate with a seat and cut off the flow of gas. When no voltage is applied, the seal is at $x_1=0$, and the spring forces F_s and membrane viscoelastic forces F_v are in equilibrium. As Maxwell force F_m increases with increasing voltage, the seal moves toward the seat to a new equilibrium position. At sufficient voltage the Maxwell force overcomes viscoelastic, frictional, and gas pressure forces so that the seal contacts the seat resulting in a reactive force at the seal-seat interface

$$
\mathbf{F}_r = \mathbf{F}_s + \mathbf{F}_m - \mathbf{F}_v - \mathbf{F}_g - \mathbf{F}_f \tag{13}
$$

The reactive force F_r , equivalent to the blocking force in [21], for a given applied voltage can be increased by designing the seat as close as possible to the seal. However, due to the long term effects of creep and fatigue the initial zero-voltage equilibrium position of the seal will drift towards the seat so that designing with the initial seal position too close to the seat would cause the valve to fail prematurely. It is clearly important to be able to predict these time-dependent effects under the given loading and temperature conditions in order to accurately design the seal to seat gap to avoid gas leakage and increase the reliability of the valve.

Potentially, an electroactive EAP membrane can be used as a sensor and modeled as a capacitor with the capacitance being a function of the actuator geometry and the dielectric properties of the polymer [22]. As the membrane extends, the capacitance will increase in a predictable and repeatable manner. With proper design of circuitry, an EAP valve can exploit this built-in displacement sensor without the need for a separate sensing device. It should be noted that since highly piezoelectric materials like PVDF have the additional property of generating voltages when deformed, this property could also be used to measure actuator position. However for static or very slow frequency measurements, determination of position

from measurement of capacitance may prove to be more accurate and will be considered here for the fault detection algorithm.

Long-term drift can be measured and compared to the predicted creep models. Specifically, and estimated damage function, $\tilde{D}(n)$, can be updated using the empirically determined model and a count of total valve cycles, n. When the valve is closed, the constant strain is known, and when the valve is open, the stress is known, based on the known force Fs and the respective position of the valve. Using the material characterization provided by the constitutive Eqs. (7) and (8) together with the measured opening and closing times and positions, it will be possible to simulate an estimated strain, $\hat{\varepsilon}(n)$ estimate the accumulated damage and compare this estimate to the measured strain giving the rule

$$
\left|\widehat{\epsilon}(n)-\epsilon(n)\right|
$$

where k denotes a certain limit based on the accuracy of the estimates, and violation of the rule signifies a valve fault. In practice, accurate simulation of $\varepsilon(n)$ may be computationally intensive. Instead, an approximation consisting of a series of exponential terms will be sought based on the analysis of equations (7) and (8). It is hypothesized that this estimator could be designed with a low order digital filter and would not be computationally intensive.

Conclusions

Experimental results show that the mechanical properties of electroactive polymers are time-dependent. Within certain limits creep deformations of EAP measured at room temperature can be accurately predicted using the constitutive equations of linear viscoelasticity. The dynamic response of EAP subjected to superimposed static and cyclic loads is characterized by accelerated creep rates due to the effects of creep-fatigue interaction. Cyclic creep acceleration has been observed even in the range of stresses well below the viscoelastic linearity limit. It is

clear that the cyclic response of the EAP is essentially nonlinear, since it does not represent a simple superposition of the responses to static and fully reversed cyclic loads applied separately.

 $\mathcal{C}^{(1)}$

To characterize cyclic damage evolution in PVDF, a constitutive material model has been developed based on the principles of linear viscoelasticity and continuum damage mechanics. The model has been validated experimentally. Fatigue induced material degradation has been characterized by experimentally determined damage function. This approach provides a reliable predictive capability for assessing the long term integrity and functionality of EAP.

The study presented in this paper provides a basis for ongoing research initiatives at the NASA Kennedy Space Center in the pursuit of new technological developments using EAP as active elements for lunar exploration systems.

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