

STOCHASTIC DELAMINATION SIMULATIONS OF NONLINEAR VISCOELASTIC COMPOSITES DURING CURE¹

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DEDICATION

This paper is dedicated to our dear friend and colleague **Dr. Y. K. Lin**, Charles E. Schmidt Professor and Director of the Center for Applied Stochastic Research, Florida Atlantic University, on the occasion of his seventy-fifth birthday.

ABSTRACT

Expressions for cure and residual stresses for nonlinear anisotropic viscoelastic composites are formulated. Probabilities of delamination during the manufacturing process are examined to determine survivability during manufacture. It is shown that the worst conditions occur during the cool down period after cure has been completed. Linear viscoelastic material maximum stresses exceed the corresponding nonlinear ones by as much as 35 %. The corresponding largest probability of failure difference is only 11 %. The effects of fiber orientation are demonstrated.

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1 INTRODUCTION

Delamination of composite plies due to interlaminar stresses at or near free edges is not necessarily to be considered an ultimate structural failure, but rather an initiation failure process which may ultimately lead to entire structural disintegration. Delamination may undergo either stable or unstable growth, with the latter leading to eventual complete failures. However, its initiation at a flight vehicle outer surface may severely damage aerodynamic performance and/or control without necessarily totally destroying its flight capabilities or terminally impairing its total structural integrity. In this paper, the word failure is used in a generic sense interchangeably with the occurrence of delamination onset.

Stochastic failure analyses must be treated on two distinct levels: (1) the analysis of stresses and displacements due to random loads, creep and relaxation functions (moduli and compliances), temperatures, moisture contents and boundary conditions; and (2) the formulation of appropriate failure criteria including proper random failure property descriptions and prescriptions. Composite materials, whether polymeric or rubber like, inherently exhibit time dependent viscoelastic responses to loads, temperatures and moisture contents and also respond with viscoelastic failure properties which degrade with time, temperatures and moistures at comparable rates, but wholly independently, both in value and mechanisms, of the degradations associated with their viscoelastic creep and relaxation functions.

The delamination problem may be divided into the following interrelated parts:

- (A) Prescription of environmental and boundary conditions
- (B) Characterization of constitutive relations and failure conditions
- (C) Determination of temperature fields
- (D) Analyses of stresses, strains and displacements
- (E) Failure (delamination) analysis

Hilton *et al.* (1991) have presented an extensive literature survey regarding effects of random loads, temperatures, humidity and material properties on viscoelastic stress analyses. Additionally, that paper also extends the traditional linear deterministic elastic-viscoelastic analogies (Christensen 1982, Hilton 1964) to analyses with stochastic material properties, but failure analyses are not covered there. Statistical data on creep and/or relaxation functions during service or manufacturing seems unavailable in the literature, but deterministic data has been extensively reported by Lazan (1968) and Nashif *et al.* (1985) and by Sierakowski & Chaturvedi (1997) for elastic composites. Stochastic delamination analyses for linear viscoelastic materials may be found in Hilton & Yi (1993, 1996a), Hilton *et al.* (1998c)

and Yi *et al.* (1996b). Detailed deterministic solutions for linear elastic composites have been covered by Vinson (1988, 1993a, b) and Vinson & Sierakowski (1987). Analytical and computational solutions for relatively simple linear viscoelastic problems are generally obtainable by analogy, if corresponding elastic solutions exist (Hilton *et al.* 1991, Christensen 1982, Hilton 1964). In more complicated problems, finite element methods (FEM) must be used and a large number of deterministic FEM are available and were compared by the present authors (Yi 1997a; Yi *et al.* 1994, 1995a, c, d, 1996b, 1997b, c, 1998c) who also have developed more computationally efficient anisotropic viscoelastic FEM for quasi-static and dynamic deterministic viscoelasticity. These FEM were used to determine stresses in composites and other anisotropic nonhomogeneous viscoelastic media. Stochastic FEM for elastic media have been developed among others by Ghanem & Spanos (1991) and Kleiber & Hien (1992).

Deterministic FEM viscoelastic solutions and protocols have been developed by Hilton *et al.* (1992, 1993, 1997a) and Yi *et al.* (1994, 1995a, c, d, 1996b, 1997b, c, 1998c). Nonlinear viscoelastic constitutive relation developments may be found in Dillard & Brinson (1983), Knauss & Losi (1993), Schapery (1969), Yi (1997a) and Yi *et al.* (1996a, 1997c). Bogetti and Gillespie (1991, 1992), Pagano & Soni (1983), Pagano (1986), Soni & Pagano (1987) and Tandon & Pagano (1994, 1996) have described and analyzed elastic delamination phenomena and edge effects.

The quadratic delamination onset criterion was first introduced by Brewer & Lagace (1988) and has recently been extended to cover time dependent creep strengths and stresses by Yi (1993). Hilton (1996b) has formulated general stochastic failure criteria under combined loads and strengths. In the present paper, the Yi delamination criterion has been generalized to cover 3-D stress states in nonlinear viscoelastic media due to random loads, temperatures, humidity, moduli and time dependent failure stresses using the stochastic combined load failure criteria of Hilton (1996b) and Hilton & Ariaratnam (1994). This formulation results in analytical relations between probabilities of failure and times at which delaminations may occur (Hilton & Yi 1993).

Very little statistical experimental delamination failure data is available in the literature. Hiel *et al.* (1991) present experimental probability distributions for interlaminar tensile strengths of curved beams and in one case examine the effects of 0% and 1.6% moisture contents. Weibull distributions are used to curve fit the data, although other distribution functions, such as Gaussian, beta, etc. (Lin 1967) could also readily be used. Other deterministic and statistical unidirectional composite time dependent failure data has been reported by Lifshitz & Rotem (1970), Phoenix & Tierney (1982), Phoenix (1979), Phoenix (1979), Phoenix & Beyerlein (1997), Watson & Smith (1985), Farquhar *et al.* (1989) and Larder & Beadle (1979). In the absence of other experimental data, one can consider that the failure density function is representable by any of the five examples given by Hiel *et al.* (1991) for deterministic temperatures and moistures.

It is very difficult to measure interlaminar stresses and particularly so during curing.

No experimental data on nonlinear viscoelastic interlaminar stresses has been reported in the literature. The model developed in this paper can be used to optimize manufacturing processes, to minimize residual stresses during cool down and to predict failure probabilities over the time range of the manufacturing process.

The present analysis was carried out for typical cure cycles based on the nonlinear viscoelastic constitutive relations developed by Schapery (1969) and Yi *et al.* (1995b-d, 1996a, 1997b, c, 1998c) and, thus, extends the authors' previous linear viscoelastic manufacturing and in-service probabilistic failure analyses.

2 ANALYSIS

2.1 Nonlinear Constitutive Relations

Following the developments of Hilton & Dong (1964b) and Hilton & Yi (1996a), these relations can be extended to large deformation behavior during manufacturing in terms of corresponding relaxation or creep functions (ϕ or ψ) in spatial intrinsic curvilinear coordinates $\theta = (\theta^i) = (\theta^1, \theta^2, \theta^3)$ and time t as

$$\begin{aligned}
\tau_j^i(\theta, t) = & \underbrace{\int_{-\infty}^t \phi_{jl}^{ik}(\theta, t, t', \Xi^{\mathbf{a}}) \frac{\partial \gamma_k^l(\theta, t')}{\partial t'} dt'}_{\text{mechanical property integrals } \mathcal{I}_j^i} \\
& - \underbrace{\int_{-\infty}^t \phi_{jl}^{ik\mathbf{T}}(\theta, t, t', \Xi^{\mathbf{a}}) \frac{\partial \hat{A}_k^l \hat{T}(\theta, t')}{\partial t'} dt'}_{\text{thermal expansion integrals } \mathcal{I}_j^{i\mathbf{T}}} \\
& - \underbrace{\int_{T^*}^T \phi_j^{i\text{th}}(\theta, t, t', \Xi^{\mathbf{b}}) dt'}_{\text{thermal change integrals } \mathcal{I}_j^{i\text{th}}} - \underbrace{\int_{\alpha^*}^{\alpha} \phi_j^{i\text{ch}}(\theta, t, t', \Xi^{\mathbf{b}}) dt'}_{\text{chemical change integrals } \mathcal{I}_j^{i\text{ch}}} \quad (1)
\end{aligned}$$

where τ_j^i and γ_j^i are large deformation stresses and strains. The material property functions Ξ are given by

$$\Xi^{\mathbf{a}} = \Xi_{jl}^{ika} [T(\theta, t', \alpha), M(\theta, t', \alpha), \alpha(\theta, t', T), \dot{I}(\theta, t')] \quad (2)$$

and

$$\Xi^{\mathbf{b}} = \Xi_j^{ib} [T(\theta, t', \alpha), M(\theta, t', \alpha), \alpha(\theta, t', T), \dot{I}(\theta, t')] \quad (3)$$

with temperature T , moisture M and degree of cure α . The strain rate invariants $\dot{I} = \{\dot{I}_1, \dot{I}_2, \dot{I}_3\}$ are defined as

$$\dot{I}_1(\theta, t) = \dot{\gamma}_i^i(\theta, t) \quad \dot{I}_2(\theta, t) = \dot{\gamma}_j^i(\theta, t) \dot{\gamma}_i^j(\theta, t) \quad \dot{I}_3(\theta, t) = \dot{\gamma}_k^i(\theta, t) \dot{\gamma}_j^k(\theta, t) \dot{\gamma}_i^j(\theta, t) \quad (4)$$

The last two integrals of Eqs. (??) are only present in the system during manufacturing cycles, where they represent changes due to cure. Dimensional changes which have been observed with time at constant temperatures \hat{T}_c and constant moisture contents are represented by the thermal integrals in Eqs. (??), provided they are recast into the form

$$\mathcal{I}_j^{i\mathbf{T}}(\theta, t) = - \int_{-\infty}^t \frac{\partial \phi_{jl}^{ik\mathbf{T}}(\theta, t, t')}{\partial t'} \hat{A}_k^l \hat{T}_c dt' \quad (5)$$

The relaxation functions ϕ_{jl}^{ik} can be represented in a number of different ways, as for instance, in terms of power series expansions (Yi *et al.* 1998a)

$$\phi_{jl}^{ik} = \phi_{jl}^{ik}(\theta, t, t', \mu, M, T, \alpha, \dot{I}) = \quad (6)$$

$$\sum_{\mathbf{m}=0}^{\underline{M}_{jl}^{ik}} \sum_{\mathbf{p}=0}^{\underline{P}_{jl}^{ik}} \sum_{\mathbf{q}=0}^{\underline{Q}_{jl}^{ik}} \left\{ B_{j\mathbf{l}\mathbf{m}\mathbf{p}\mathbf{q}}^{ik}(\theta, t, t', M, T, \alpha) (\dot{I}_1 - 3)^{\mathbf{m}} (\dot{I}_2 - 3)^{\mathbf{p}} (\dot{I}_3 - 1)^{\mathbf{q}} \right\}$$

where underscores indicate no summations over the affected indices and all other parameters are material dependent. The parameter μ is a measure of aging effects (Brinson & Gates 1995). Additional forms of aging models may be found developed and summarized in Drozdov (1998) and their representations can be readily incorporated into Eqs. (??) and (??).

Alternately, when expressing material characterization in term of generalized Kelvin or Maxwell models, relaxation functions may be expressed as nonlinear Prony series with relaxation times $\hat{\tau}_{jln}^{ik}$ as

$$\phi_{jl}^{ik}(\theta, t, t', \mu, M, T, \alpha, \dot{I}) = \quad (7)$$

$$\phi_{j\infty}^{ik} + \sum_{\mathbf{n}=0}^{\underline{N}_{jl}^{ik}} \phi_{j\mathbf{l}\mathbf{n}}^{ik}(\theta, M, T, \alpha, I) \exp \left[- \int_{\underline{\tau}_{j\mathbf{l}\mathbf{n}}^{ik}(\theta, t', M, T, \alpha, \dot{I})}^t dt' \right]$$

Schapery (1969) has devised an approximate practical isotropic nonlinear constitutive relation which has been extended by Yi *et al.* (1996a) to nonlinear anisotropic viscoelasticity under multiaxial loads as

$$\tau_j^i(\theta, t) = \quad (8)$$

$$\sum_{k=1}^9 \sum_{l=1}^9 \{ h_{ijkl}^{\infty} \phi_{ijkl}^{\infty} \gamma_{kl}(\theta, 0) + h_{ijk\mathbf{l}T}^{\infty} \phi_{ijk\mathbf{l}T}^{\infty} \alpha_{kl} T(\theta, 0) \}$$

$$\begin{aligned}
& + h_{ijkl}^1 \int_0^t \phi_{ijkl}(\theta, t, t') \frac{\partial [h_{ijkl}^2 \gamma_{kl}(\theta, t')]}{\partial t'} dt' \\
& - h_{ijklT}^1 \int_0^t \phi_{ijklT}(\theta, t, t') \frac{\partial [h_{ijklT}^2 \alpha_{kl} T(\theta, t')]}{\partial t'} dt'
\end{aligned}$$

Yi (1997a) and Yi *et al.* (1996a, 1998c) have used this constitutive formulation previously in non-manufacturing problems and results of material responses based on this characterization can be found in these references.

2.2 Cure Cycle Temperature

Fortunately, the heat balance relations governing the determination of temperatures during cure and cool down cycles are uncoupled from those governing stresses, strains and displacements even for nonlinear constitutive relations. Consequently, the temperature field is determined by governing relations for curing epoxy resins which are well established and will only be briefly listed here. The heat of reaction H at any time t is given by

$$H(\theta, t) = \int_0^t \frac{1}{\rho} \left(\frac{dQ}{ds} \right) ds \quad \text{and} \quad H_T(\theta, t_f) = \int_0^{t_f} \frac{1}{\rho} \left(\frac{dQ}{ds} \right) ds \quad (9)$$

where $\rho(\theta, t) = \rho(\theta, t, \alpha, T)$ is the composite density, $dQ(\theta, s, T)/ds$ the heat generation rate per unit volume and t_f the final time required to complete the reaction with an amount H_T . The degree of cure α is defined by

$$\alpha[x, t, T(\theta, t)] = \frac{H(x, t)}{H_T(\theta, t_f)} \quad \text{with} \quad 0 \leq \alpha \leq 1 \quad (10)$$

which then gives the governing equation for α as

$$\frac{d\alpha}{dt} = F(\theta, t, \alpha, T) \quad (11)$$

where $T = T(\theta, t, \alpha)$ is the temperature. The function F is material property dependent and examples are listed in Yi *et al.* (1998a, b).

From Fourier's conduction law for an anisotropic nonhomogeneous thermal conductivity matrix $\mathbf{k}(\theta, t) = \mathbf{k}(\theta, t, \alpha, T)$, one obtains from the energy balance over the entire volume

$$\int_V \underbrace{\left[\rho(c - H_T) \frac{\partial T}{\partial t} + \nabla(\mathbf{k} \nabla T) \right]}_{=0} dV = 0 \quad (12)$$

with $c(\theta, t, \alpha, T)$ the specific heat and thus requiring the integrand to vanish throughout the volume V . The initial and boundary conditions are respectively

$$T(\theta, 0) = T^0(\theta) \quad (13)$$

$$\bar{a}_1 \frac{\partial T}{\partial \mathbf{n}} + \bar{a}_2 T + \bar{a}_3 T_s = \bar{a}_4 \quad \text{for } T_s(\theta, t) \text{ on } \Gamma(\theta) \quad (14)$$

where $\mathbf{n}(\theta)$ is the direction normal to the boundary surface $\Gamma(\theta)$ enclosing the body volume $V(\theta)$, while the quantities $\bar{a}_i(x, t, \alpha, T)$ are material dependent heat transfer coefficients on Γ .

It is evident that Eqs. (??) and (??) are coupled and must be solved simultaneously as has been demonstrated by Yi *et al.* (1998a), among others, using FEA / FDA and typical results are shown in Yi *et al.* (1998a, b). Fig. 1 shows cure temperatures and pressures in the autoclave during the manufacturing cycle.

2.3 Multiaxial Failure Simulations

Hilton & Ariaratnam (1994) have formulated an empirical invariant combined stress failure law, which is applicable to both deterministic and stochastic conditions. These probabilistic formulations have been applied to column creep delamination buckling and to linear viscoelastic service and manufacturing delamination onset predictions (Hilton *et al.* 1993, 1996a, b, 1997a, b). Hiel *al.* (1991) have experimentally shown that uniaxial composite delamination failures obey Weibull distributions (Lin 1967).

Consider applied stress invariants defined by

$$\tilde{\mathcal{J}}_1 = \tilde{\tau}_i^i \quad \tilde{\mathcal{J}}_2 = \tilde{\tau}_i^j \tilde{\tau}_j^i \quad \tilde{\mathcal{J}}_3 = \tilde{\tau}_i^j \tilde{\tau}_i^k \tilde{\tau}_k^j \quad (15)$$

with similar expressions for the uniaxial failure stresses F_{ij}

$$\tilde{\mathcal{F}}_1 = \tilde{F}_i^i \quad \tilde{\mathcal{F}}_2 = \tilde{F}_i^j \tilde{F}_j^i \quad \tilde{\mathcal{F}}_3 = \tilde{F}_j^i \tilde{F}_i^k \tilde{F}_k^j \quad (16)$$

where the functions with a \sim indicate stochastic quantities and those without are their corresponding mean values. Mean uniaxial viscoelastic delamination strengths are shown in Fig. 2.

The failure criteria are now cast into

$$\frac{1}{3} \sum_{i=1}^3 \left(\frac{\tilde{\mathcal{J}}_i}{\mathcal{J}_i} \right)^{c_i} = \tilde{V} \quad \text{and} \quad \frac{1}{3} \sum_{i=1}^3 \left(\frac{\tilde{\mathcal{F}}_i}{\mathcal{F}_i} \right)^{c_i} = \tilde{v} \quad (17)$$

Failure occurs whenever

$$\tilde{u} = \tilde{V} - \tilde{v} \geq 0 \quad (18)$$

Uniaxial failure data by Hiel *et al.* (1991) for epoxy/fiber composite delaminations indicate Weibull distributions (Hilton & Yi 1993, Lin 1986), consequently for combined loads the failure probability then becomes

$$\tilde{P}(t) = 1 - \exp \left\{ - \left[\frac{\tilde{u}(t)}{\beta} \right]^A \right\} \quad (19)$$

where \mathcal{A} and β are experimentally determined material parameters.

3 DISCUSSION

T300/5208 graphite/epoxy composites are considered for the numerical simulations. Using creep and creep recovery tests, the master compliance curves and shift factors corresponding to various loading conditions were obtained from Tuttle and Brinson (1985). The orthotropic viscoelastic material can be described by the usual short hand notation of classifying relaxation functions with six indices ($\phi_{11}^{11} \equiv \phi_{11}$, etc.). The simulation defines time functions for ϕ_{33} equal to ϕ_{22} and $\phi_{66} = \phi_{44} = \phi_{55}$. The relaxation function ϕ_{11} is taken to be elastic because it is generally controlled by fiber properties. Under these conditions, Poisson's ratio ν_{12} is constant and ϕ_{12} has the same time function as ϕ_{22} . The time independence requirements of isotropic and anisotropic Poisson ratios were derived by Hilton & Yi (1998). The laminate width to thickness ratio is four and the ply thickness d is 1.4224×10^{-4} m.

The thermophysical properties considered during this simulation in Eq. (??) are $\mathbf{k}_{33} = \mathbf{k}_{22} = 0.4135 \text{ W/m}^\circ\text{K}$, $\mathbf{k}_{11} = 2.0675 \text{ W/m}^\circ\text{K}$, $\rho = 1,520 \text{ Kg/m}^3$ and $c = 942 \text{ J/Kg}^\circ\text{C}$. Due to lack of experimental data, the the thermal conductivities \mathbf{k} are taken as anisotropic, but temperature independent. The effects of anisotropic \mathbf{k} s on stresses during cure in linear anisotropic viscoelastic materials are discussed by Yi *et al.* (1996b).

T300/5208 laminates with $(0_{10}/90_{10})_s$ and $(45_{10}/-45_{10})_s$ layups are considered and nine node isoparametric elements were used. The finite element model consists of a 56×16 mesh in the yz cross-section with a total of 11,187 degrees of freedom. The step-size Δt is set to 60 secs initially and Δt increases with time. There are 30 time steps involved in the calculation of time-dependent interlaminar stresses over a period of 1×10^5 secs.

Fig. 3 shows distributions of shear stresses τ_{xz} in the width direction of the plate at various times. Clearly, the free edge stresses at $y = \pm .015$ increase drastically with time as the cool down process takes place reaching their largest value at the end of cool down at 3480 sec after cure stops. Figs. 4 and 5 depict the various free edge stress components for $(0_{10}/90_{10})_s$ and $(45_{10}/-45_{10})_s$ for nonlinear viscoelastic material configurations. Note that while all stress components increase with time and the pattern changes with fiber orientations since for $(0_{10}/90_{10})_s$ the normal stresses τ_{zz} are the largest tensors, while for $(45_{10}/-45_{10})_s$ τ_{xz} predominates. Fig. 6 is comparable to Fig. 5, except that the viscoelastic material is linear and produces considerably larger stresses. This phenomenon is demonstrated again in Fig. 7 where it is shown that the maximum difference between linear and nonlinear stress reaches 35 % at the end of the cool down cycle, thus making the linear stress analysis results extremely conservative. Fig. 8 displays the probability of delamination as a function of cool down time for linear and nonlinear materials for these two fiber orientations. However, the probability of delamination which takes into consideration the combined effects of all stress components differs only by 11 % for the linear and nonlinear 45° laminates.

The analysis shows that the one dimensional loading produces responses with a three dimensional stress field from which stress invariants can be applied using the invariant failure

theory developed by Hilton & Ariaratnam (1994). The worst conditions occur during cool down near the end of cycle when residual stresses are highest and failure strengths have had sufficient time to degrade. It is seen that there are some configurations where delaminations are predicted with probabilities approaching unity resulting in certain failures during manufacture and thus making the cured composite unacceptable for service. Other configurations exhibit sufficiently high failure probabilities to render the manufactured product of equally unacceptable quality. These manufacturing failure problems can be alleviated by shortening the cure cycle, using materials with slower degradation rates or reducing cure temperatures and pressures. No current materials seem to exist which admit lower cure temperatures and pressures and/or shorter cure manufacturing cycles. Hilton & Yi (1992) and Beldica & Hilton (1998) have formulated isotropic and anisotropic analytical designer materials for a number of service conditions, but the inverse problem of how to safely and effectively manufacture such composites has yet to be formulated and analyzed.

4 CONCLUSIONS

For the cure manufacturing process, the largest stresses occur during cool down cycles. However, because of changing moduli and failure stresses with temperature, moisture, degree of cure α , strain invariants, etc., this does not necessarily preclude delamination prior to cool down when $0 \leq \alpha \leq 1$ and $t \leq 300$ min. In the present simulation, the severest failure conditions and, consequently, the highest probabilities of failure occur at the end of the cool down period at 3480 sec after curing terminated. Different fiber orientations produce distinct residual stress patterns and probabilities of delaminations. Large differences (35 %) were observed between stresses for linear and nonlinear viscoelastic materials, but the nonlinear influence on corresponding failure probabilities yields only one third as large a difference.

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Fig. 1 - CURE TEMPERATURE & PRESSURE

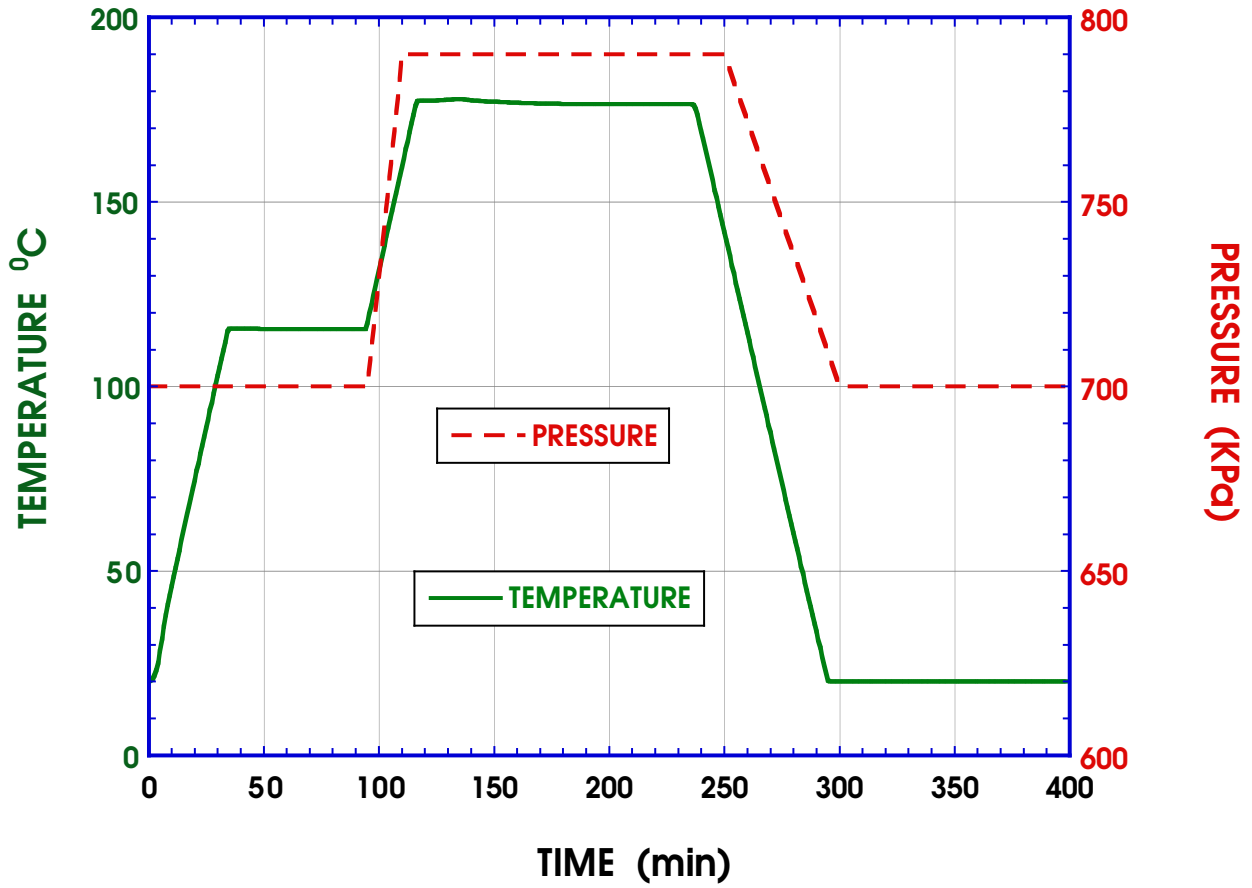
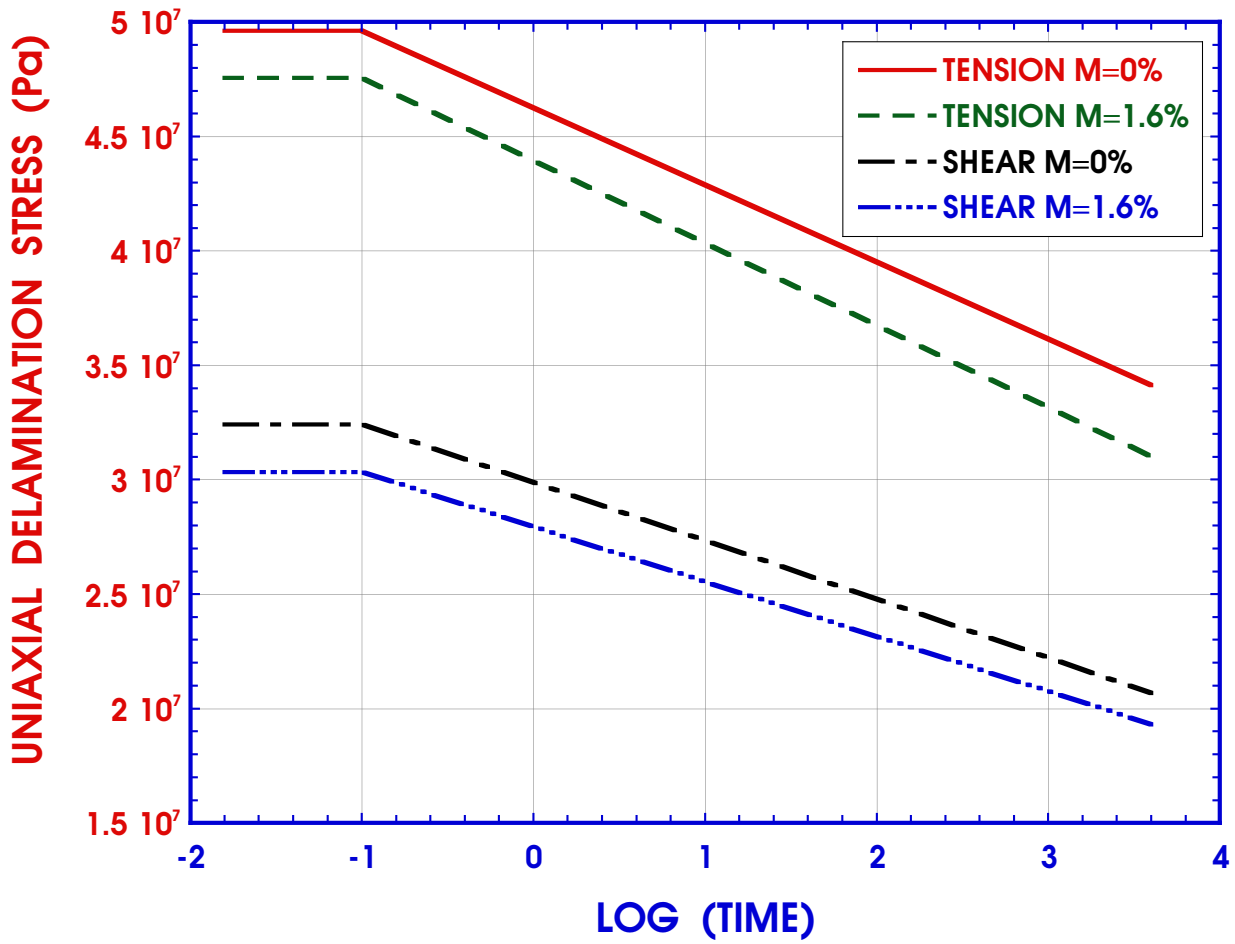


Fig. 2 - DELAMINATION UNIAXIAL CREEP STRENGTH



**Fig. 3 - NONLINEAR INTERLAMINAR SHEAR STRESS
(45₁₀/-45₁₀)_S DURING COOLING PROCESS**

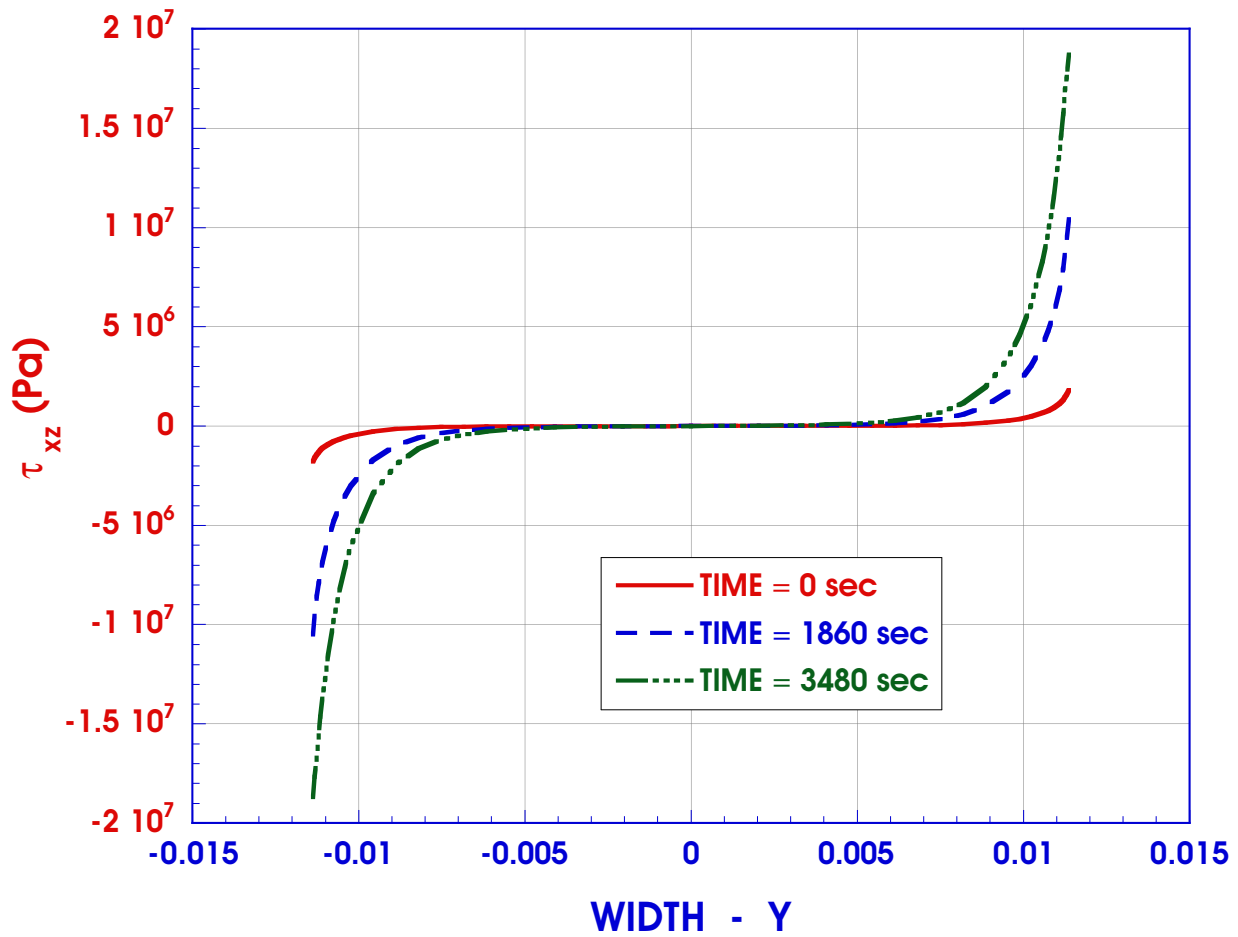


Fig. 4 - NONLINEAR EDGE STRESSES FOR $(0_{10}/90_{10})_s$

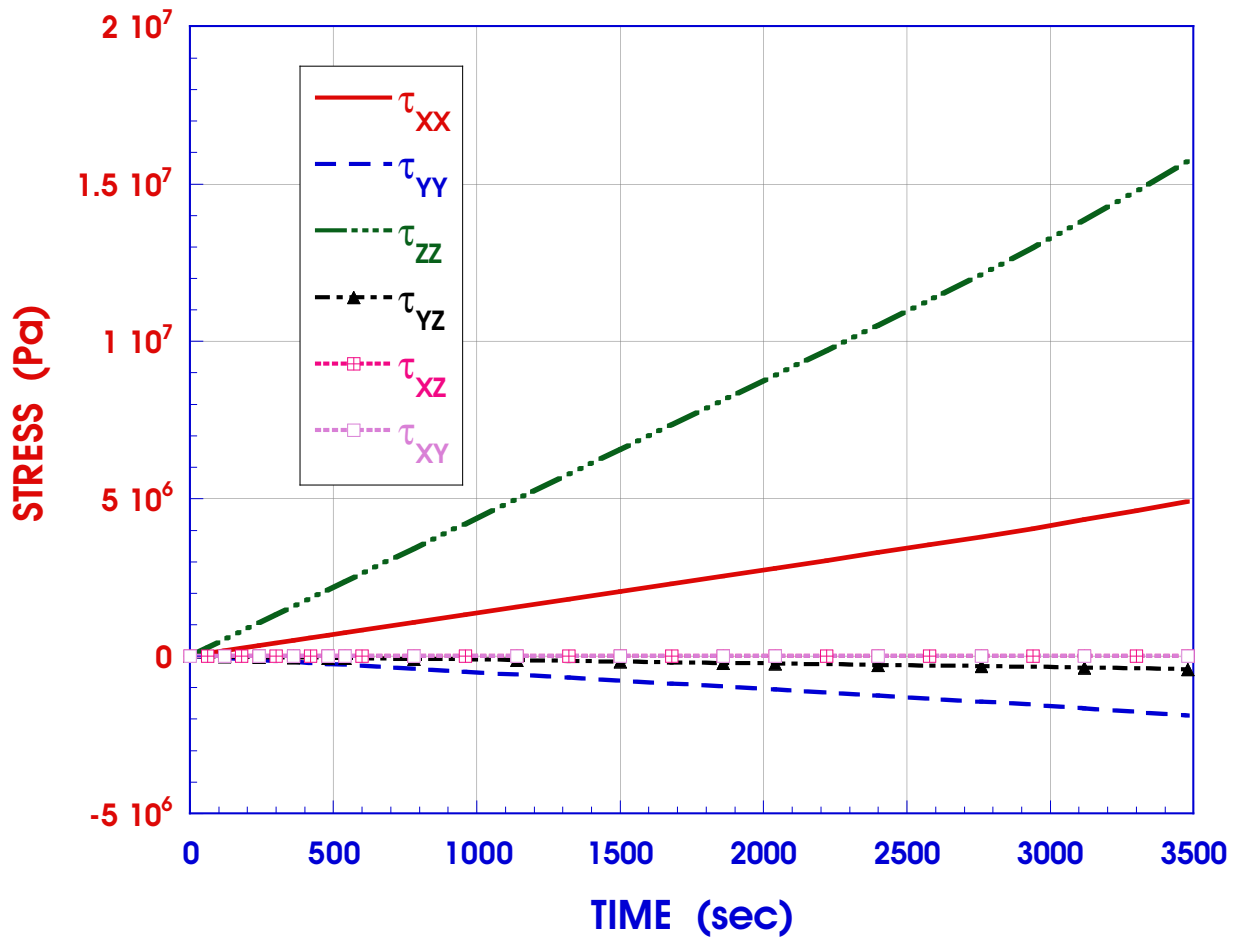


Fig. 5 - NONLINEAR EDGE STRESS FOR $(45_{10}/-45_{10})_s$

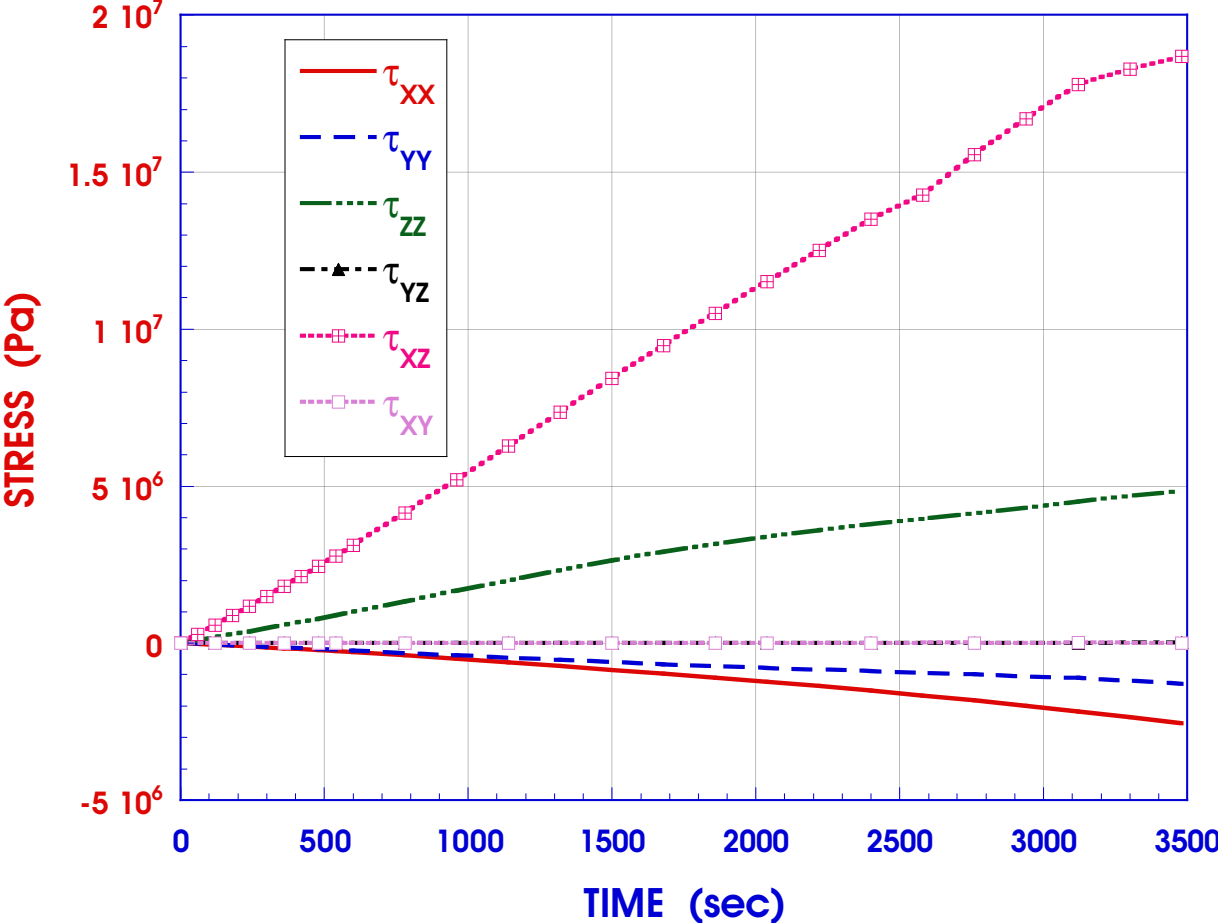


Fig. 6 - LINEAR EDGE STRESSES FOR $(45_{10}/-45_{10})_S$

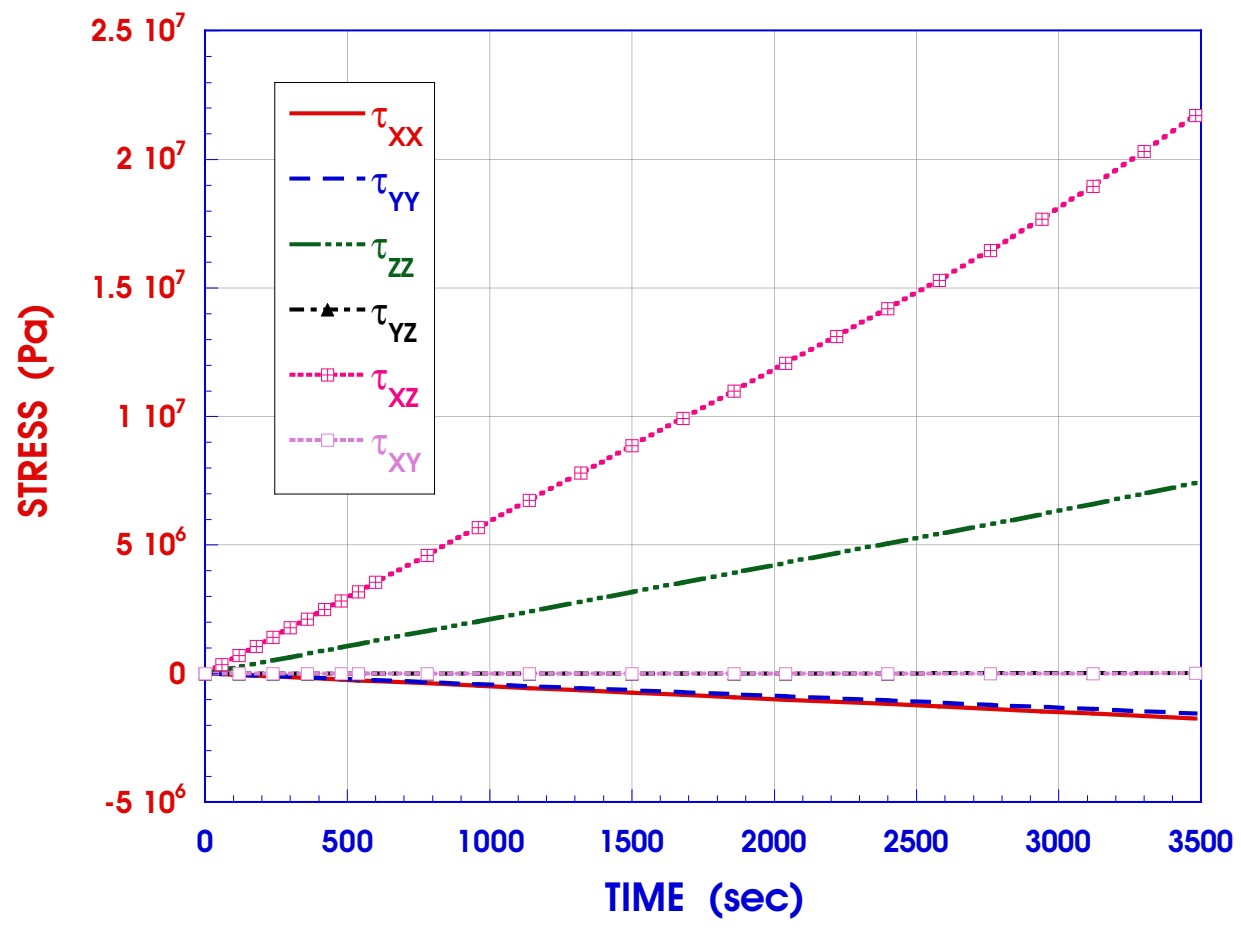


Fig. 7 - LINEAR & NONLINEAR EDGE STRESSES σ_{zz}
 FOR $(45_{10}/-45_{10})_s$ LAMINATES

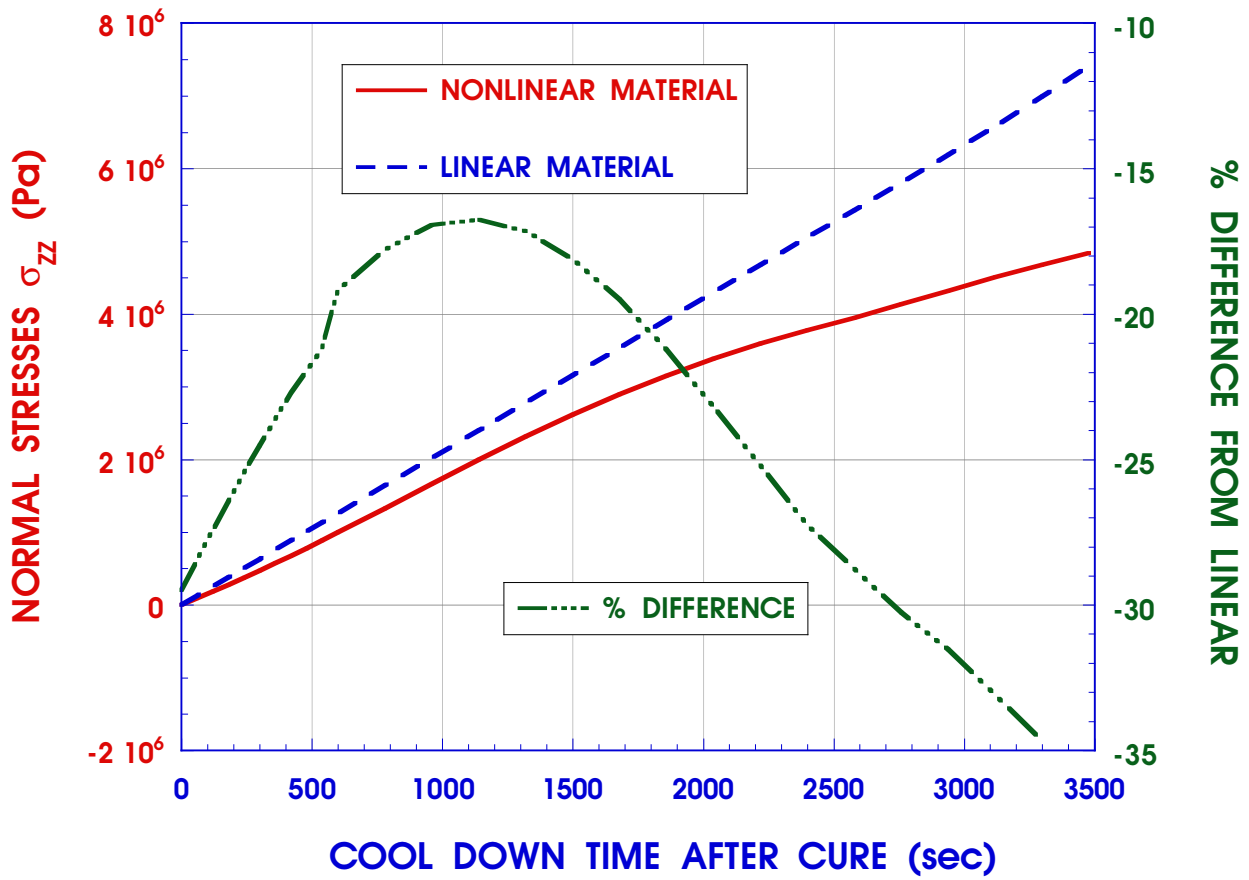


FIG. 8 - PROBABILITY OF DELAMINATION ONSET DURING COOL DOWN

