THERMO-MECHANCAL CHANGES OF PLASTICS UNDER ENVIRONMENTAL CHANGES

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Abstract

In this paper, Polypropylene composites, a popular thermo-plastic material for automobile instrument panels, were characterized for mechanical property changes under hygro-thermal accelerated test conditions to investigate the temperature and humidity effects on the Young's modulus and Poisson's ratio changes by aging. And the data were modeled based on exponential functions to predict long term properties change. It was found that the Young's modulus was increased as the thermal aging was continued, while the moisture effects were not noticeable in the aging time range considered in this study. The property changes model was suggested by maximum likelihood method, where uncertainties were taken into account due to the measurement error and lack of data.

1 Introduction

The mechanical behaviours of engineering plastics are significantly influenced by environmental changes. For automobile applications, the plastic structures for dashboard, instruments and other panels are continuously exposed to sun lights and moisture, which lead to substantial changes of material properties. The physical and mechanical property changes of polymeric materials are due to their unique amorphous molecular structure, and categorized as physical aging and degradation [1, 2]. Many literatures are available on the aging phenomena, and it is known that the behaviour changes stem from thermodynamic equilibrium process. The memory effects under Tg is erased and recovered (rejuvenated) when the material experiences the temperature above Tg [3-8]. On the other hand, degradation is not well defined and commonly used as same as physical aging. However, degradation distinguishes itself as irreversible process due to permanent changes of the molecular structures, which may be induced by moisture, oxygen, ultraviolet light and chemical attacks. In the material characterizations, however, it is not easy task to separate these two phenomena from each other [9-11]. The analyses of the behavior changes become essential, particularly in the interior design of automobiles. Perceivable interior noises such as buzz, squeak and rattle (BSR) are important issues in the design of the automobile industry. Mostly due to small gaps or preload in the assembled polymer parts, they are also created by aging and degradation of engineering plastics, which render the loss of structural integrities. Therefore, to consider BSR from initial stage of the interior design, it is very important to obtain, analyze and understand the structural behaviors of the materials under environmental changes as well as time. In this study, the engineering plastic material property changes were measured under accelerated temperature and humidity changes with different degrees. The material properties of Young's modulus and Poisson's ratio were periodically measured to investigate the properties change. For the long term predictions, a model was also established based on the data by considering uncertainties by the measurement errors and lack of data.

2 Materials and test data

2.1 Materials

Two Polypropylene (PPF) composites named as Type A and Type D for automobile globe box were selected for the degradation tests. According to ASTM D5229/D5229M-92 procedure D, all the specimens were desiccated in vacuum oven at 60°C during 8 hours. The desiccated specimens are homogenized on 23 ± 2 °C, relative humidity 50%±5% R.H. for 40 hours. Then the specimens were aged under three different conditions for isothermal degradation and two conditions for moisture absorption according to SAE J1455. For the isothermal degradation, 85 °C, 75 °C and 65 °C were chosen. For the moisture absorptions, 85% relative humidity and 45% relative humidity at 85°C were employed. Then the specimens were periodically taken out to measure the material properties.

2.2 Tests under Isothermal Degradation Environments

Young's modulus and Poisson's ratio were measured and the variations are presented in Fig. 1. Due to the nonlinear stress-strain curve, the modulus was measured by 1% and 2% secant modulus. As seen, Young's modulus of both polymers increased in the early stage of the tests, and remained unchanged, then increased again. Meanwhile, the Poisson's ratio was increased in the initial stage, then slightly decreased. The results indicate that the modulus does not necessarily decrease due to the environmental aging.

2.3 Tests under Moisture Absorption Environments

Moisture absorption tests consist of two conditions 45% and 85% humidity at 85 $^{\circ}$ C. Again, 1% and 2% secant modulus were measured and the results are represented in Fig. 2. As seen, no significant degradation of the modulus changes were found during the 70 days, which was against usual expectation of the adverse effects of the moisture.. For the case of Poisson's ratio, the data show an increase in the initial stage, and the changes are small afterward.







Figure 1. Tensile property variations under isothermal conditions.







Figure 2. Tensile property variations under moisture absorption.

3 Properties model

3.1 Model selection

For the modeling, an exponential function was assumed for the time effects such as [13, 14]

$$\mu(t) = a \exp(bt) \tag{1}$$

For the temperature and humidity, an Arrhenius degradation model was selected and expressed as

$$\mu(T) = a \exp(b/T) \tag{2}$$

Eq.(1) and Eq.(2) can be transformed into a single model

$$\ln(\mu(t)) = \alpha - \beta' t \tag{3}$$

$$\beta' = \beta \exp(-\gamma / T) \tag{4}$$

Or as the final combined form as

$$\ln\left(\mu(t,T)\right) = \alpha - t\beta \exp\left(-\gamma/T\right)$$
(5)

where *T* is to represent temperature and humidity and α , β , γ are variable to be estimated. *3.2 Maximum Likelihood Estimation*

In this paper, maximum likelihood estimation (MLE) is used to estimate α , β , γ in Eq. (5) based on the measured data. MLE point-estimates model parameters θ in a probability density function (PDF) $f(x,\theta)$ by maximizing the likelihood function $L(\theta)$, which is defined by multiplication of the PDF at each measured data. This is schematically explained in Fig. 3. Assuming that the error between the measurement and the model follows normal distribution with unknown standard deviation, the likelihood function is given as follows [15, 16].

$$L = f(x_1, \theta) \cdot f(x_2, \theta) \cdots f(x_n, \theta) = \prod_{i=1}^n \frac{1}{\sqrt{2\pi\sigma}} \exp\left[-\frac{1}{2}\left(\frac{\ln x_i - \mu_i}{\sigma}\right)^2\right]$$
(6)

where x_i and μ_i denote the measured data and model value at *i* th measurement condition respectively, and σ is the standard deviation of the error. Note that the model μ is given by Eq. (5). Then the set of unknown parameters θ consist of α , β , γ and σ . The problem is now to determine θ that maximizes the function *L*. In practical computation, log likelihood is employed as follows.

$$\ln L = -n \ln \sqrt{2\pi} - n \ln \sigma - \frac{1}{2} \sum_{i=1}^{n} \left(\frac{\ln x_{i} - \mu}{\sigma} \right)^{2}$$

$$\ln L = -n \ln \sqrt{2\pi} - n \ln \sigma - \frac{1}{2} \sum_{i=1}^{n} \left(\frac{\ln x_{i} - (\alpha - t\beta \exp(-\gamma/T))}{\sigma} \right)^{2}$$
(7)
Random Variable
$$X_{n} = f(x_{n}, \theta)$$
Estimate θ
Maximum
Likelihood
Estimator
 $\hat{\theta}$
Likelihood function
$$L(\theta) = f(x_{1}, \theta) \cdot f(x_{2}, \theta) \cdots f(x_{n}, \theta)$$

Figure 3. Maximum Likelihood Estimation

Optimization is conducted using MATLAB to determine α , β , γ and σ of the material type A and type D using the measured data up to the 70th days. In Fig. 4, the degradation models for 1% secant modulus are also plotted using the MLE parameters along with the measured data for the two materials.





Figure 4. Degradation model of 1% secant modulus

3.3 Bootstrap Confidence Interval

Confidence interval is estimated by bootstrap method. The idea is to simulate the repeated sampling process and use the information from the distribution of appropriate statistics in the bootstrap samples to compute the confidence bounds. There are two methods for generating samples, which are nonparametric and fully parametric bootstrap sampling. When the number of samples is very small (say, less than 7 like this study), fully parametric approach is preferable. The process of this method is given in Fig. 5 and also explained as follows.

- 1) Using the measured data $X = (x_1, x_2 \cdots x_n)$, point-estimate parameters θ using MLE method.
- 2) Simulate samples of measured data using the estimated parameters θ and pointestimate parameters θ using the MLE method. This is repeated *B* times to obtain bootstrap samples of θ .
- 3) Obtain the confidence interval $\left[\theta^*(\alpha/2 \cdot B), \theta^*((1-\alpha/2) \cdot B)\right]$ from the bootstrap samples [17].



Figure 5. Fully parametric bootstrap sampling

3.4 Validation of the model

To validate the degradation model, the properties prediction is made using the model was compared with the actual measured data of the 100th day. The result for the A type isothermal 1% secant modulus is given in Fig. 6, in which the 5 number of measured data at the 100th day are enclosed within the confidence interval of the degradation model, demonstrating the model successfully predicted the properties change. Results of the other cases also fall within their confidence interval prediction, which are not included for brevity.



Figure 6. Validation of degradation model for Type A 1% secant modulus

4 Conclusions

In this paper, the material property modeling under the aging was presented. It wqas shown that the Young's modulus were increased as the thermal aging time was increased. Poisson's ratio was also increased, although the value change was not that noticeable as much as the Young's modulus. Similar trends were also found in the moisture absorption, indicating that the moisture did not cause any adverse effect on the materials used in this study. The prediction modeling based on the maximum likelihood estimation and the bootstrap confidence interval technique was successfully applied to predict the properties change. The Young's modulus changes during the 100 days considered in this study showed gradual increase of the values. We expect eventually saturated value after long time aging, and plan for further tests of the isothermal and humidity agings for extended time.

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