IEA SHC Task 27: Adoption of General Methodology to Durability Assessment of Polymeric Glazing Materials

Gary Jorgensen
National Renewable Energy Laborator, NREL
Golden, CO 80401-3393, USA

Polymeric Glazing Materials R&D

Polymeric glazings offer significant potential for cost savings both as direct substitutes for glass cover plates in traditional collector systems and as an integral part of all-polymeric systems. This savings includes both base material cost and costs associated with shipping, handling and installation (due to their lightweight and lack of fragility). Glazings should have high transmittance across the solar spectrum and must be able to resist long term (10-20 years) exposure to service conditions including elevated operating temperatures (55-90°C) and solar ultraviolet (UV) light. They must retain mechanical integrity (for example, impact resistance and flexural rigidity) under these harsh environmental stresses. Recent efforts have emphasized 1) identification of new/improved candidate glazings, 2) evaluation of optical and mechanical durability during exposure to actual and simulated service conditions, and 3) adoption of a general methodology of accelerated life testing to the durability assessment of polymeric glazing materials.

Sample Selection and Testing

A number of candidate materials have been identified by reviewing the literature and through discussions with experts within the polymer and solar manufacturing industries. Polycarbonate (PC) has high optical clarity and excellent impact strength. However, under UV exposure it will yellow and become brittle. Recently, stabilized versions of polycarbonate have been developed. For example, Bayer has two products designated APEC 5391 and APEC 5393. The first is a thermally stabilized formulation (having a maximum continuous use temperature up to 180ºC) and the second is both heat and UV stabilized. GE Plastics has incorporated an integral UV-screening coating (that is also mar-resistant) into a number of their Lexan products.

Samples of candidate polymeric glazing materials were subjected to in-service outdoor and accelerated laboratory exposure conditions. Outdoor testing was carried out in Switzerland at the Institut für Solartechnik (SPF), Germany (at ISE in Freiburg), and at three sites in the United States (Golden, CO; Phoenix, AZ; and Miami, FL). A precise and detailed knowledge of the specific environmental stress conditions experienced by weathered samples is needed to allow understanding of site-specific performance losses and to permit service lifetime prediction of candidate glazings. Consequently, each operational exposure site is fully equipped with appropriate meteorological and radiometric instrumentation and data-logging capability.

Outdoor Exposure Testing

The materials tested are for the intended use in solar thermal flat plate collectors. Thus, the samples for outdoor exposure were fixed onto mini collector boxes (see Figure 1). In order to simulate the elevated temperature collector covers are exposed to, the 'mini collectors' are made of solar selective coated stainless steel.

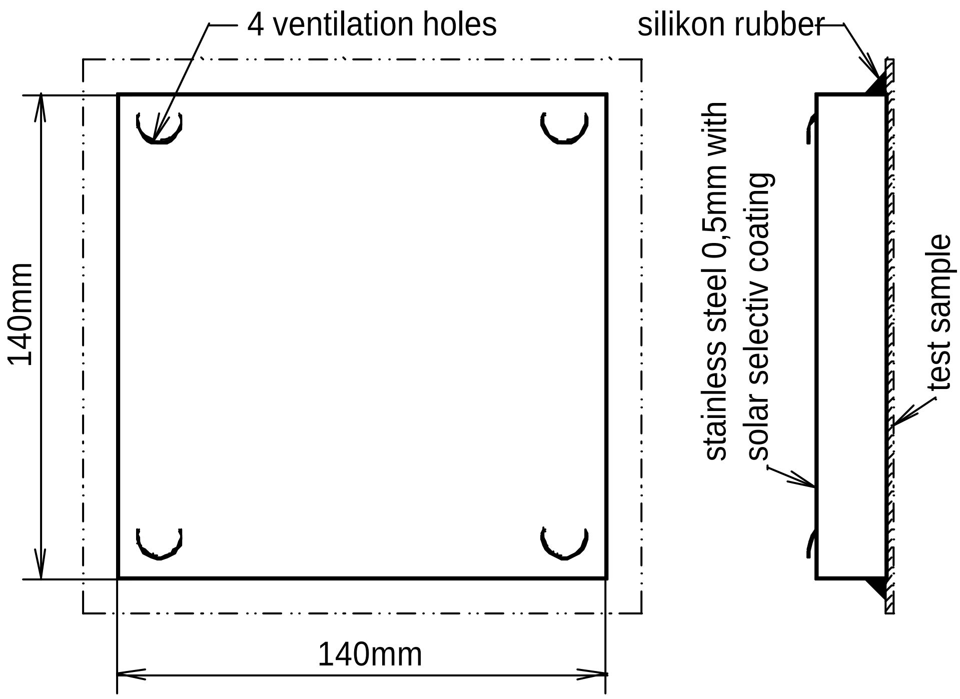

Figure 1 Mini collectors' used for outdoor exposure of transparent cover materials

The samples prepared in this way were exposed to the ambient climate at locations in Europe and in USA at an inclination angle between 45° and 60° facing south. The spectral transmittance of all samples was measured prior to exposure. Periodically, the samples were re-measured and exposed again without any cleaning. Other samples were measured before and after cleaning and then exposed again.

Accelerated Laboratory Exposure Testing

Accelerated indoor testing was carried out with the different test equipment of the participating laboratories. The following tests were performed:

- temperature (dark) test at 40ºC, 60ºC, 65ºC, 70ºC, 75ºC, and 80ºC. i.e. only elevated temperature without any irradiation

- UV test in combination with elevated temperature and a defined level of humidity at 60°C / 80% r.h., 80°C / 40% r.h. and 50°C / 95% r.h. These tests were performed in climatic cabinets with an unfiltered metal halide (HMI) lamp as the light source. The intensity of the irradiation in comparison to the AM1.5 solar spectrum is about 3 times as much UVA and 7 times as much UVB.

- Atlas Ci5000 Weather-Ometer test with irradiation close to 2x AM1.5 solar spectrum. Tests were performed with and without an additional UV filter (Korad acrylic film) affixed to the samples.

- Atlas XR35 Weather-Ometer – SPART14 test
  The SPART 14 test procedure was originally developed for clear coats in automotive paint systems. The test is a weather ability test, which includes acidic rain spraying. The test method SPART 14 is a modification of SAE J1960:
  Light source: Xenon arc, filtered through borosilicate filters
  Irradiance level: 0.5 W/m^2 at 340nm
  Test cycle: 40 min light only
  20 min light + front spray of water; in every 14th cycle the water is acidic
  with a pH of 3.2
  60 min light only
  60 min dark + back spray
  Black standard temperature and relative humidity during light periods are 70°C and 75%, respectively. Air temperature during light periods is about 47°C. During light periods the temperature inside the weather-Ometer is controlled by the black standard temperature, hence the air temperature can vary slightly.
  Temperature and relative humidity during dark periods are 38°C and 95%, respectively.
  During light periods the temperature of the tested glazing materials was estimated to reach 50-55°C
  The test specimens were exposed for up to 4000 hours. An exposure time of 1000 hours in the accelerated Weather-Ometer test is estimated to correspond to about 1.3 years of outdoor testing in Miami, Florida of automotive paints. Consequently, 4000 hours of SPART 14 testing correspond to about 5 years outdoors in Florida. However, one can assume that the temperature of an automotive coating will be at least 10K higher than for transparent low light absorbing glazing materials. Consequently, the acceleration factor for the glazing can be estimated to be a factor of 2 higher. Accordingly, 1000 hors artificial weathering correspond to 2.5 years outdoors and 4000 hours to 10 years.

Parallel testing with relevant stress factors at different levels was intended to allow the sensitivity of materials degradation to these factors to be quantified, and allow damage function models to be evaluated. This in turn can be used to compare the time-dependent performance of these materials with measured results from in-service outdoor exposure.

Highly accelerated exposure testing of selected samples was also performed at NREL using a unique UV concentrator [1]. It consists of an array of faceted mirrors that tracks the sun in two axes and redirects sunlight back to a sample exposure chamber attached by three structural support tubes. The concentrator is designed to provide up to 100X concentration having uniform flux at high UV intensity and low visible (VIS) and near-infrared (NIR) intensity. This is achieved by coating the facets with a custom-designed 37-layer film that uses alternating high and low refractive index materials that results in high UV reflectance and low VIS/NIR reflectance.

Application of General Methodology to Data Analysis

Analysis of test results was carried out as outlined in [2]. To obtain correlations between in-use and accelerated exposure results, a suitable material-specific damage function model must be found that accurately relates changes in an appropriate response variable to relevant applied environmental stresses. The response variable can be either microscopic (e.g., changes in chemical structure) or macroscopic (e.g., loss of gloss), but ideally should be easily measured and directly related to an important property of the material being tested.

Organic materials are known to be susceptible to degradation caused by cumulative light dosage (D) exposure [3]:

$$D(t) \sim \int_{0}^{t} L(t)\,dt \qquad (1)$$

where L(t) is the time-dependent incident spectral irradiance, I(λ,t), convoluted with the absorption spectra of the material being exposed, α(λ,t), and the quantum efficiency of the absorbed photons to propagate reactions that are harmful to the coating, ϕ(λ,t), integrated over an appropriate bandwidth (defined by λ_{min} and λ_{max}) throughout which light-induced damage occurs:

$$L(t) = \int_{l_{min}}^{l_{max}} I(l,t)\,a(l,t)\,f(l,t)\,dl \qquad (2)$$

In previous work [4,5] with organic materials (back-metallized polymeric films), useful results have been obtained by approximating the absorption spectra and quantum efficiency as constants in eq 2 and defining:

$$I_{UV}(t) = \int_{l_{min}}^{l_{max}} I(l,t)\;dl \qquad (3)$$

with λ_{min} = 285 nm and λ_{max} = 315 nm (for UV-B) or λ_{min} = 290 nm and λ_{max} = 385 nm (for total UV). For constant (controlled) irradiance, this leads to an approximate generalized cumulative dosage model in which the loss in performance, ΔP, (change in response variable) with time is proportional to a power law expression of the ultraviolet irradiance I_{UV} [6,7]:

$$\frac{\Delta P}{\Delta t} \sim (I_{UV})^{n} \qquad (4)$$

To account for thermal effects, an Arrhenius term can be included and the change in performance after the ith time interval is:

$$\Delta P_i = A\,(I_{UV})^{n}\;\Delta t_i\;e^{-E/kT} \qquad (5)$$

where T is the temperature (K) experienced by samples during exposure, k is Boltzmann's constant, and E is an activation energy. For constant accelerated stresses, I_{UV} and T are known; this allows eq 5 to be fit to measured values of ΔP_i and subsequent determination of the coefficients A, E, and n. For variable real-world stresses, the time dependent form eq 5 must be used:

$$\Delta P(t) = A \int_0^t \left[I_{UV}(t) \right]^n \, e^{-E/kT(t)} \, dt \qquad (6)$$

Having determined the relevant coefficients from AET's performed at constant stresses, eq 6 can be used to compute a predicted loss in performance after some time t where the relevant stresses are monitored; these predicted values can then be compared with actual measured values.

Validation of Methodology

If measured values of ΔP_i for samples of materials exposed outdoors, where the time dependent stress variables are known, then, eq 6 can be used to predict ΔP_i for comparison with measured data. As discussed in Part I, such information is available for some materials exposed at the various instrumented international outdoor test sites. To validate the methodology presented in Part III, two types of sheet (0.32-cm thick) glazing materials tested at a variety of outdoor exposure test (OET) sites will be considered. These include polyvinyl chloride (PVC) and a UV-stabilized polycarbonate (PC). These materials have also been exposed [8] in an Atlas Ci-5000 WOM (having a UV intensity of about 2X compared to typical outdoor terrestrial levels) and at 50X and 100X at NREL's UV concentrator [1]. The response variable was chosen to be hemispherical transmittance between 400-500 nm because, in general, that is the spectral region most sensitive to stress exposure induced loss in performance (Figure 2). The same damage functions expressed in eqs 5 and 6 were assumed. Data from the Ci-5000 and the UV concentrator exposures were used to fit eq 5 and to obtain the model coefficients; the results are given in Table 1. Values of activation energies (E) derived are reasonable for photo-thermally driven degradation mechanisms. A value of n ~ 2/3 for PVC implies that exposure to 50-100X light intensities had a net effect of only 15-25X, suggesting that some shielding or rate limiting reactions occur that do not allow all photons to participate in degradation. For the UV-stabilized PC sample, a value of n=1 suggests that exposure of this material follows strict reciprocity even up to 100X; all incident photons fully contribute to degradation reactions that proceed at twice the rate undergone at 50X exposure and 50 times the rate experienced at 2X exposure.

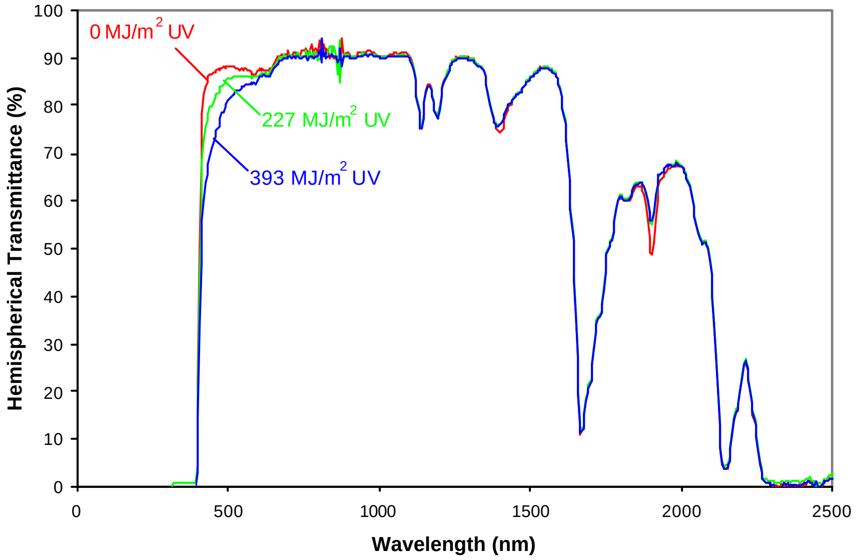

Figure 2 Change in hemispherical transmittance as a function of cumulative UV dose for UV-stabalized polycarbonate

Table 1 Coefficients Derived for Representative Clear Polymer Sheet Samples

Polymer Sheet	***A***	***N***	***E (kcal/mole-K)***
Polyvinyl Chloride	2892	0.669	8.440
UV-Stabilized Polycarbonate	5.497	1.093	6.688

Using the coefficients from Table 1 and time-monitored values of sample temperature and UV irradiance, the loss in performance was predicted using eq 6 for both PVC and PC as exposed outdoors in Golden, CO, and Phoenix, AZ. Predicted values were then compared with actual measured data for these materials exposed at these sites. The results are presented in Figure 3. Time-dependent changes in weathering variables produce the irregular shapes of the predicted curves. Excellent agreement is evident between the measured and predicted data, thereby validating the ability to expose samples at very high light levels, our approach to data analysis (using accelerated test results to obtain model coefficients, and then the use of these coefficients to predict time-variable real-world degradation), and the assumed damage function model.

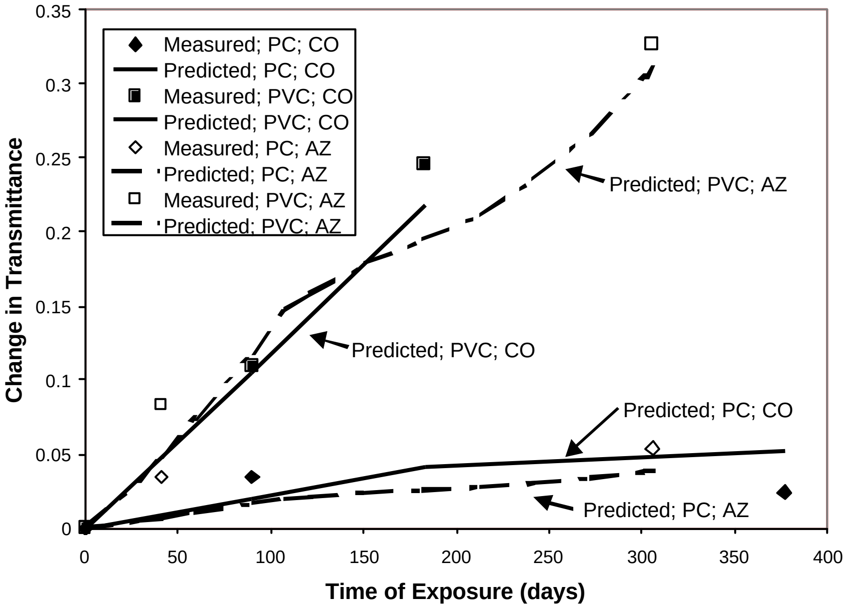

Figure 3 Measured vs. predicted change in hemispherical transmittance between 400-500 nm for two polymeric materials at two outdoor exposure sites.

Conclusions

A large amount of durability test data for both accelerated laboratory conditions and outdoor in-service conditions has been acquired for PC and PVC glazing materials. Some of this accelerated exposure data has been used to demonstrate how to derive damage functions that allow prediction of performance degradation. This methodology also allows the effect of multiple stress factors to be modeled. The usefulness and validity of this approach has then been confirmed by comparing predicted results with actual measured data for samples exposed to variable outdoor conditions. Consequently, very abbreviated testing times at elevated stress conditions can be substituted for long-time exposures at lower stress levels. This will allow much shorter development cycle times for new products and will allow improvements to be identified and readily incorporated.

References

1. Jorgensen, G.; Bingham, C.; King, D.; Lewandowski, A.; Netter, J.; Terwilliger, K.; and Adamsons, K. In *Service Life Prediction Methodology and Metrologies*; Bauer, D. R. and Martin, J. W., Eds.; ACS Symposium Series xxx; American Chemical Society, Oxford University Press: Washington, DC, 2001; pp yyy-zzz (in press).

2. General methodology of accelerated testing for assessment of service life of materials; Bo Carlsson, SP Swedish National Testing and Research Institute; Working document of IEA Task 27.

3. Martin, J. W.; Saunders, S. C.; Floyd, F. L.; Weinberg, J. P. *Methodologies for Predicting the Service Life of Coatings Systems*, NIST Building Science Series 172, National Institute of Standards and Technology: Gaithersburg, MD, 1994.

4. Jorgensen, G.; Bingham, C.; Netter, J.; Goggin, R.; Lewandowski, A. In *Service Life Prediction of Organic Coatings, A Systems Approach*; Bauer, D. R. and Martin, J. W., Eds.; ACS Symposium Series 722; American Chemical Society, Oxford University Press: Washington, DC, 1999; pp 170-185.

5. Jorgensen, G.; Kim, H-M.; Wendelin, T.; In *Durability Testing of Nonmetallic Materials*; Herling, R. J., Ed.; ASTM STP 1294; American Society for Testing and Materials: Scranton, PA, 1996; pp 121-135.

6. Martin, J. W. Durability of Building Materials **1982**, 1, 175-194.

7. Carlsson, B. Solar Materials Research and Development; Survey of Service Life Prediction Methods for Materials in Solar Heating and Cooling; International Energy Agency Solar Heating and Cooling Program, Report # BFR-D-16-1989 (DE90 748556), ISBN 91-540-5063-4; Swedish Council for Building Research: Stockholm, Sweden, 1989; p 27.

8. Raman, R. et al, “A Review of Polymer Materials for Solar Water Heating Systems”, Journal of Solar Energy Engineering, Vol. 122, No. 2, May 2000, pp. 92-100.